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# TITAN: A Trajectory-Informed Technique for Adaptive Parameter Freezing in Large-Scale VQE

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## 1 Appendix

### 2 A Related Works

#### 3 A.1 (i) Measurement–Cost Mitigation

4 **Commuting–set and graph–based grouping.** Early works exploited qubit–wise commuting  
5 (QWC) partitions, typically found via minimum–clique–cover heuristics on Pauli–term graphs. While  
6 grouping can slash shot counts, the NP–hard clique search still scales poorly and demands one distinct  
7 circuit per group. Recent “overlapped–grouping” frameworks generalise QWC by allowing *shared*  
8 observables between groups; the resulting convex allocation of shots outperforms prior heuristics  
9 across 16-qubit molecular Hamiltonians[1].

10 **Classical–shadow variants.** Classical shadows randomise measurement bases and reconstruct  
11 many expectation values simultaneously, but uniform sampling wastes shots on insignificant terms.  
12 Derandomised and importance–weighted shadows address this by pre–computing tailored bases, at  
13 the cost of *classical* post–processing that scales quadratically in the number of measured terms[2, 3].  
14 The very recent ROGS algorithm blends overlapped grouping with shadow tomography and optimally  
15 allocates a fixed shot budget via a confidence–bound objective, lowering *both* shot *and* unique–circuit  
16 counts on benchmarks up to 20 qubits[4].

17 **Learning–based grouping.** Graph neural networks and reinforcement learning have been used to  
18 predict near–optimal partitions directly from Pauli–Hamiltonian structure[5]. These methods amortise  
19 the combinatorial search after training, but require sizeable data sets of Hamiltonians and incur  
20 non-negligible offline costs.

21 Measurement strategies trade quantum shots for classical runtime; none alter the *optimization*  
22 landscape itself, leaving barren-plateau issues untouched.

#### 23 A.2 (ii) Ansatz Design and Compression

24 **Hardware-efficient vs. problem-inspired circuits.** Hardware-efficient ansätze (HEA) minimise  
25 depth on specific devices but often suffer from barren plateaus (BPs)[6]. In contrast, chemically  
26 or symmetry inspired ansätze (e.g., UCC or symmetry-preserving layouts) improve trainability yet  
27 dramatically increase two-qubit-gate count[7].

28 **Adaptive and search-based construction.** ADAPT-VQE builds circuits iteratively by adding  
29 operators with the largest energy gradient, offering compact, system-specific wavefunctions but  
30 requiring hundreds of rounds of gradient evaluations [8]. Architecture-search frameworks, both

evolutionary[9] and Bayesian[10], scan supercircuit spaces to identify expressive, shallow layouts, yet introduce an outer optimization loop whose cost grows combinatorially with qubit number.

**Pruning and parameter sparsification.** Once trained, many ansätze contain redundant gates. Gradient-based pruning[11] and Hessian-guided methods such as QADAPRUNE[12] delete low-saliency parameters on the fly, reducing depth without re-training. The *Pruned-ADAPT-VQE* refinement removes near-zero-amplitude operators from adaptive circuits, shrinking molecular ansätze by up to 30 %[13].

Circuit-level improvements fight either expressibility or depth; they do not reduce the *dimension* of the parameter space fed to the classical optimizer.

### A.3 (iii) Advanced Classical Optimizers

**Quantum-aware gradients.** Natural-gradient and quantum-Fisher techniques exploit the information-geometry of parameterised unitaries but double (or worse) the number of hardware evaluations per step. QBANG approximates the Fisher matrix via a Broyden update and mixes it with momentum, matching natural-gradient accuracy with gradient-descent cost[14].

**Meta-learning and hyper-opt.** Meta-optimizers learn update rules across VQE tasks[15] and can warm-start large-scale instances, yet inject their own nested objective and hyperparameter. RL-based schedulers dynamically switch between gradient-free and gradient-based steps, but still operate in the *full* parameter space.

**Noise-aware and Bayesian schemes.** Techniques that re-weight gradients by empirical shot noise or adopt Bayesian optimization have improved robustness on real hardware, though at the price of surrogate-model maintenance and additional variance estimates[16].

**Take-away.** Optimizer innovations reshape the search trajectory yet remain bottlenecked by high-dimensional parameter landscapes.

### A.4 (iv) Parameter-Space Reduction

A complementary axis—*shrinking the search space itself*. Early works froze parameters whose gradients fall below a threshold, but fixed thresholds risk misclassifying temporarily stagnant yet ultimately useful parameters [17]. TITAN extends this idea by predicting freezing *before training* via a CFCSA encoder and APFA labels, reducing trainable parameters and measurement shots simultaneously. Unlike pruning (which acts *after* or *during* optimization), predictive freezing is *proactive*, feeding a leaner search space to *any* optimizer or measurement scheme.

**Synergy with prior axes.** Because freezing removes variables rather than adding gates or shots, TITAN layers orthogonally atop (i)–(iii): fewer parameters lower the classical cost of natural-gradient estimators, shorten architecture-search evaluations, and even tighten confidence bounds in shadow-based measurement allocation.

## B APFA Implementations Details

The APFA mechanism dynamically identifies and *freezes* low-saliency parameters in a VQE to record the freezing trajectory as shown in Figure 1. For a VQE with parameter vector  $\theta^t \in \mathbb{R}^P$  at iteration  $t$ , we define the stochastic gradient

$$\mathbf{g}_t = \nabla_{\theta} f(\theta^t) + \xi_t, \quad \xi_t \sim \mathcal{N}(\mathbf{0}, \gamma^2 \mathbf{I}), \quad (1)$$

where  $\xi^t$  is an isotropic noise term (with scale  $\gamma$ ) designed to improve exploration. APFA *dynamically* toggles each parameter  $\theta_i$  ( $i = 1, \dots, P$ ) between an *active* and a *frozen* state, according to the four steps below:

**Gradients and Exponential Moving Average (EMA)** To quantify parameter *saliency*, an exponential moving average (EMA) of the absolute gradient is maintained for each coordinate  $i \in \{1, \dots, P\}$ :

$$\hat{g}_t^{(i)} = \alpha \hat{g}_{t-1}^{(i)} + (1 - \alpha) |g_t^{(i)}|, \quad 0 < \alpha < 1, \quad (2)$$

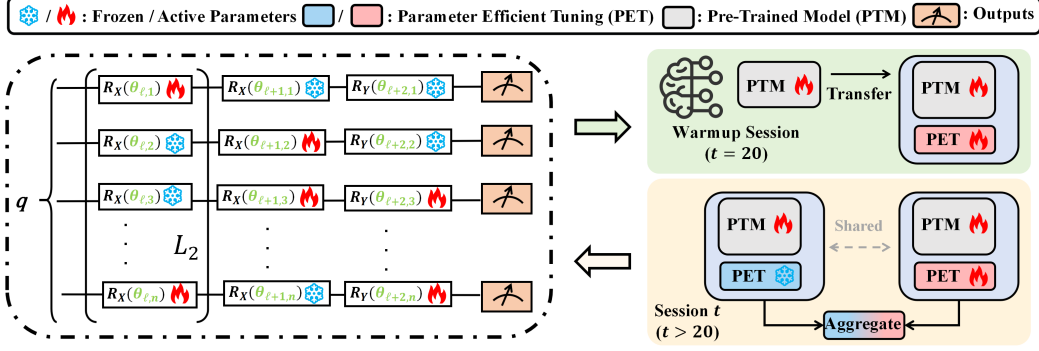


Figure 1: An illustration of the TITAN architecture employing APFA to freeze or reactivate parameters. The red flames mark active (trainable) parameters, while blue snowflakes indicate frozen coordinates.

with  $\hat{g}_0^{(i)} = |g_0^{(i)}|$  and  $g_t^{(i)}$  the  $i$ th component of  $\mathbf{g}_t$ . This EMA captures recent trends in gradient magnitudes while smoothing out short-term fluctuations.

**Global Magnitude and Dynamic Thresholding** We track two global metrics at iteration  $t$ . First, the mean of the EMA of absolute gradients,  $\bar{g}_t = \frac{1}{P} \sum_{i=1}^P \hat{g}_t^{(i)}$ , provides a representative measure of overall gradient magnitude. Second, we compute a *decay ratio*  $r_t$  defined by

$$r_t = \frac{\|\mathbf{g}_t\|_2}{\|\mathbf{g}_0\|_2 + \varepsilon}, \quad \varepsilon \approx 10^{-12}, \quad (3)$$

which compares the current global gradient norm to its initial value. This ratio serves as an adaptive control signal for scaling the *freeze* factor  $\lambda_f^{(t)}$  and *activate* factor  $\lambda_a^{(t)}$ :

$$\lambda_f^{(t)} = \begin{cases} 3\lambda_f^{(0)}, & r_t < 0.2, \\ \lambda_f^{(0)}[1 + 2(1 - r_t)/0.8], & r_t \geq 0.2, \end{cases} \quad \lambda_a^{(t)} = \begin{cases} 3\lambda_a^{(0)}, & r_t < 0.2, \\ \lambda_a^{(0)}[1 + 2(1 - r_t)/0.8], & r_t \geq 0.2. \end{cases} \quad (4)$$

Thus, if  $r_t$  becomes sufficiently small (i.e., the global gradient norm decays sharply), the factors  $\lambda_f^{(t)}$  and  $\lambda_a^{(t)}$  both increase, encouraging more aggressive freezing and reactivation thresholds.

The resulting *freeze* and *activate* thresholds are given by  $\tau_f^{(t)} = \lambda_f^{(t)} \bar{g}_t$ ,  $\tau_a^{(t)} = \lambda_a^{(t)} \tau_f^{(t)}$ . Therefore, any coordinate whose EMA magnitude  $\hat{g}_t^{(i)}$  falls below  $\tau_f^{(t)}$  repeatedly is considered low-saliency and may be *frozen*, while coordinates rising above  $\tau_a^{(t)}$  sufficiently often are *reactivated*.

**State Machine for Freezing and Activating Parameters** To implement the above thresholds, we maintain two counters,  $c_f^{(i)}$  and  $c_a^{(i)}$ , and a binary mask  $m_t^{(i)} \in \{0, 1\}$  for each coordinate  $i$ . If  $m_t^{(i)} = 0$ , the parameter is *frozen*, whereas if  $m_t^{(i)} = 1$ , it is *active*. The counters record how many consecutive iterations  $\hat{g}_t^{(i)}$  has stayed below  $\tau_f^{(t)}$  (for freezing) or exceeded  $\tau_a^{(t)}$  (for activating). Formally, for user-defined integers  $N_f$  (freeze) and  $N_a$  (activate), the update rule is:

$$\begin{aligned} \text{if } m_{t-1}^{(i)} = 1 \text{ (active):} & \quad \begin{cases} c_f^{(i)} \leftarrow c_f^{(i)} + 1, & \text{if } \hat{g}_t^{(i)} < \tau_f^{(t)}, \\ c_f^{(i)} \leftarrow 0, & \text{otherwise,} \end{cases} \quad \text{freeze if } c_f^{(i)} \geq N_f; \\ \text{if } m_{t-1}^{(i)} = 0 \text{ (frozen):} & \quad \begin{cases} c_a^{(i)} \leftarrow c_a^{(i)} + 1, & \text{if } \hat{g}_t^{(i)} > \tau_a^{(t)}, \\ c_a^{(i)} \leftarrow 0, & \text{otherwise,} \end{cases} \quad \text{activate if } c_a^{(i)} \geq N_a. \end{aligned} \quad (5)$$

As soon as a counter  $c_f^{(i)}$  or  $c_a^{(i)}$  crosses its respective threshold  $N_f$  or  $N_a$ , we update  $m_t^{(i)}$  to 0 or 1 (i.e., freeze or activate). Letting  $\mathbf{m}_t$  denote the vector of masks at iteration  $t$ , a generic first-order optimizer subsequently performs the following masked parameter update:

$$\theta_{t+1} = \theta_t - \eta_t (\mathbf{m}_t \odot \mathbf{g}_t), \quad (6)$$

where  $\eta_t$  is the learning rate and  $\odot$  denotes the Hadamard product. Only active parameters (i.e., coordinates with  $m_t^{(i)} = 1$ ) are updated, while frozen ones remain fixed.

**Convergence and Practical Considerations** Under standard smoothness and bounded-below assumptions on  $\mathcal{L}$  (i.e.,  $\mathcal{L}$  being  $L$ -smooth and coercive), with learning rate sequence  $\{\eta_t\}$  satisfying  $\sum_t \eta_t = \infty$ ,  $\sum_t \eta_t^2 < \infty$ , and  $\lambda_f^{(t)} \rightarrow 0$  no faster than  $\eta_t$ , the analysis in [18] can be adapted to show  $\liminf_{t \rightarrow \infty} \|\nabla \mathcal{L}(\theta_t)\|_2 = 0$ , i.e., APFA retains the standard stochastic gradient convergence guarantees. In practice, once parameters are frozen, the optimization is effectively running in a reduced-dimensional subspace, which can lead to faster progress on significant coordinates while still allowing reactivation when needed. Consequently, this *parameter-efficient* approach can offer both computational savings and improved generalization performance by reducing overfitting on low-saliency dimensions.

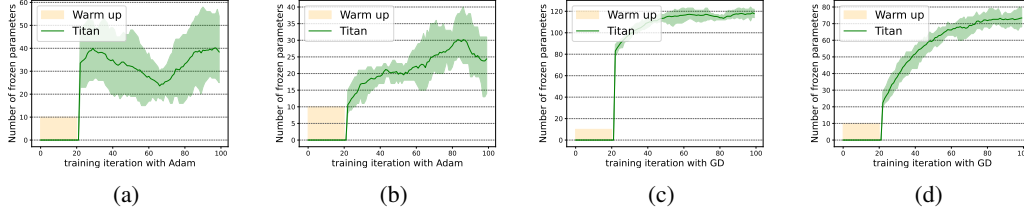


Figure 2: Evolution of the number of frozen parameters under different optimizers and noise settings of the Heisenberg model (HEA, XX+YY+ZZ, Qubits: 10, Layers: 15) (a) Adam without noise. (b) Adam with noise. (c) Gradient Descent without noise. (d) Gradient Descent with noise.

Figure 2 tracks the cumulative number of gates identified as redundant by APFA a 10-qubit, 15-layer HEA circuit for the  $XX + YY + ZZ$  Heisenberg Hamiltonian. The yellow band marks the *warm-up phase* ( $t \leq T_{\text{warm}}$ ), whereas the green curve (with  $\pm\sigma$  envelope,  $n = 5$  seeds) shows the ensuing Titan-mediated freezing trajectory. Overall, Titan adapts its freezing rate to the optimizer-noise landscape: stochastic Adam benefits from *incremental* freezes that hedge against noisy gradients, whereas deterministic GD permits *aggressive* early pruning.

## C Proof for Theorem 1

For convenience, we list two useful lemmas before the proof of Theorem 1.

**lemma 1.** Consider the unitary  $U$  sampled from the unitary 2-design in  $\mathcal{U}(2)$ . Let  $A, B \in \{I, X, Y, Z\}$ . Then we have

$$\mathbb{E}_U (U^\dagger A U) \otimes (U^\dagger B U) = \begin{cases} I \otimes I, & \text{when } A = B = I, \\ \frac{1}{3} (X \otimes X + Y \otimes Y + Z \otimes Z), & \text{when } A = B = X/Y/Z, \\ 0, & \text{for other cases.} \end{cases} \quad (7)$$

*Proof.* Lemma 1 can be obtained by using Eq. (53) in Ref. [19] for  $A, B \in \{I, X, Y, Z\}$ .

□

**lemma 2.** Consider the function  $f = \text{Tr}[(O_1 \otimes O_2)(R \otimes A)B(R^\dagger \otimes A^\dagger)]$ , where  $R = \exp[-i\theta G/2]$  and the Hamiltonian  $G$  is either commuting or anti-commuting with  $O_1$ . Then

$$\mathbb{E}_\theta f^2 = \begin{cases} (\text{Tr}[(O_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)])^2, & \text{when } GO_1 = O_1G, \\ \frac{1 + \exp[-2\gamma^2]}{2} (\text{Tr}[(O_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)])^2 \\ + \frac{1 - \exp[-2\gamma^2]}{2} (\text{Tr}[(iGO_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)])^2, & \text{when } GO_1 = -O_1G, \end{cases}$$

119

$$\mathbb{E}_{\theta} \left( \frac{\partial f}{\partial \theta} \right)^2 = \begin{cases} 0, & \text{when } GO_1 = O_1 G, \\ \frac{1 - \exp[-2\gamma^2]}{2} (\text{Tr} [(O_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)])^2 \\ + \frac{1 + \exp[-2\gamma^2]}{2} (\text{Tr} [(iGO_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)])^2, & \text{when } GO_1 = -O_1 G, \end{cases}$$

120 where  $\theta$  is sampled from  $\mathcal{N}(0, \gamma^2)$ .

121 *Proof.* For the case of  $GO_1 = O_1 G$ , the result yields trivially. We focus on the case of  $GO_1 =$   
 122  $-O_1 G$ , where the function  $f$  has the format

$$\begin{aligned} f &= \text{Tr} [(O_1 \otimes O_2)(\exp[-i\theta G/2] \otimes I)(I \otimes A)B(I \otimes A^\dagger)(\exp[i\theta G/2] \otimes I)] \\ &= \text{Tr} [(\exp[i\theta G] \otimes I)(O_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)] \\ &= \cos \theta \text{Tr} [(O_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)] + \sin \theta \text{Tr} [(iGO_1 \otimes O_2)(I \otimes A)B(I \otimes A^\dagger)]. \end{aligned}$$

123 By calculating the expectation of  $\cos^2 \theta$ ,  $\sin^2 \theta$ , and  $\sin \theta \cos \theta$ , we obtain Lemma 2.

124

□

125 **Theorem 3** (Enhanced Gaussian Initialization). Consider the VQC  $V(\theta) =$   
 126  $(\otimes_{n=1}^N U_n) \prod_{i=1}^L \text{CZ}_i \text{RY}_i(\theta^{2i-1}) \text{RX}_i(\theta^{2i})$ , where each  $U_n$  is sampled from unitary 2-design  
 127 in  $\mathcal{U}(2)$  and each parameter in  $\theta$  is sampled from  $\mathcal{N}(0, \gamma^2)$  independently with  $\gamma \leq 1$ . Consider the  
 128  $S$ -local observable with the Pauli decomposition  $O = \sum_{\mathbf{i}} \alpha_{\mathbf{i}} P_{\mathbf{i}}$ . Let  $f = \text{Tr}[OV(\theta)|0\rangle\langle 0|V(\theta)^\dagger]$  be  
 129 the loss function. Then for each partial derivative  $\frac{\partial f}{\partial \theta^{m,n}}$ , the expected squared gradient (with respect  
 130 to both the random unitaries  $U = \otimes_{n=1}^N U_n$  and the random parameters  $\theta$ ) admits the following  
 131 lower bound:

$$\mathbb{E}_{U, \theta} \left( \frac{\partial f}{\partial \theta^{m,n}} \right)^2 \geq \frac{1}{3^S} \sum_{\mathbf{i}} \alpha_{\mathbf{i}}^2 (1 - \delta_{i_n 0}) (1 - \gamma^2)^{2LS} \frac{2}{3} \gamma^2, \quad (8)$$

132 where  $S = \max_{\mathbf{i}} (\|\mathbf{i}\|_0)$ . Moreover, choosing  $\gamma^2 = c/L$  ensures that

$$\mathbb{E}_{U, \theta} \|\nabla_{\theta} f\|^2 \geq \mathcal{O}(1) \quad (9)$$

133 for the case of local observable  $O = \sum_{\mathbf{i}} \alpha_{\mathbf{i}} P_{\mathbf{i}}$  with  $S = \mathcal{O}(1)$  and  $\alpha_{\mathbf{i}} = \mathcal{O}(1)$ .134 *Proof.* For convenience, we denote intermediate states

$$\rho_k := \left( \prod_{i=L+1-k}^L \text{CZ}_i \text{RY}_i(\theta^{2i-1}) \text{RX}_i(\theta^{2i}) \right) \rho_0 \left( \prod_{i=L+1-k}^L \text{CZ}_i \text{RY}_i(\theta^{2i-1}) \text{RX}_i(\theta^{2i}) \right)^\dagger, \quad (10)$$

135 where  $\rho_0 = |0\rangle\langle 0|$ . Therefore, we can rewrite the loss function as

$$f(\theta) = \text{Tr} [OU \rho_L(\theta) U^\dagger], \quad (11)$$

136 where  $U = \otimes_{n=1}^N U_n$  is the final single-qubit unitary layer. Based on the parameter-shift rule, the  
 137 partial derivative has the format

$$\begin{aligned} \frac{\partial f}{\partial \theta^{m,n}} &= \frac{1}{2} [f(\theta_+) - f(\theta_-)] \\ &= \frac{1}{2} [\text{Tr} [OU \rho_L(\theta_+) U^\dagger] - \text{Tr} [OU \rho_L(\theta_-) U^\dagger]] \\ &:= \text{Tr} [OU \rho'_L U^\dagger], \end{aligned} \quad (12)$$

138 where  $\theta_+$  and  $\theta_-$  differ from  $\theta$  by adding  $\pi/2$  or  $-\pi/2$  on the term  $\theta^{m,n}$ , and  $\rho'_L := \frac{\rho_L(\theta_+) - \rho_L(\theta_-)}{2}$ .

139 Next, we consider the expectation of the square of the partial derivative term under the distribution of  
 140  $U$ . By using the Pauli decomposition of the observable  $O = \sum_i \alpha_i P_i$ , we have

$$\begin{aligned} \mathbb{E}_U \left( \frac{\partial f}{\partial \theta^{m,n}} \right)^2 &= \mathbb{E}_U (\text{Tr} [OU \rho'_L U^\dagger])^2 \\ &= \sum_{i,j} \alpha_i \alpha_j \mathbb{E}_U \text{Tr} [P_i U \rho'_L U^\dagger] \text{Tr} [P_j U \rho'_L U^\dagger] \end{aligned} \quad (13)$$

$$= \sum_i \alpha_i^2 \frac{1}{3^{\|\mathbf{i}\|_0}} \sum_{j \in \text{Supp}(\mathbf{i})} \text{Tr} [P_j \rho'_L]^2 \quad (14)$$

$$\geq \frac{1}{3^S} \sum_i \alpha_i^2 \text{Tr} [Z_i \rho'_L]^2, \quad (15)$$

where  $\text{Supp}(\mathbf{i})$  denotes the set

$$\{(k_1, k_2, \dots, k_N) | k_n \in \{1, 2, 3\} \text{ if } i_n \neq 0; k_n = 0 \text{ if } i_n = 0\},$$

141 and Eq. (14) is derived by using Lemma 1. Eq. (15) is derived since each Pauli basis in  $O$  with non-  
 142 zero coefficient has at most  $S$  non-identity elements ( $S$ -local). The term  $Z_i$  denotes the observable  
 143 consisted of  $Z$  and  $I$ , where  $Z$  is applied when the corresponding coefficient in  $\mathbf{i}$  is not zero.

144 Next, we proceed to the analysis of the expectation of Eq. (15) with respect to  $\theta$ . Let  $j := \lfloor \frac{m+1}{2} \rfloor$ .  
 145 For the case of  $j > 1$ , we have

$$\begin{aligned} \mathbb{E}_{\theta^1, \theta^2} \text{Tr} [Z_i \rho'_L]^2 &= \mathbb{E}_{\theta^1, \theta^2} \text{Tr} \left[ Z_i \text{CZ}_1 \text{RY}_1(\theta^1) \text{RX}_1(\theta^2) \rho'_{L-1} \text{RX}_1(\theta^2)^\dagger \text{RY}_1(\theta^1)^\dagger \text{CZ}_1^\dagger \right]^2 \\ &= \mathbb{E}_{\theta^1, \theta^2} \text{Tr} \left[ Z_i \text{RY}_1(\theta^1) \text{RX}_1(\theta^2) \rho'_{L-1} \text{RX}_1(\theta^2)^\dagger \text{RY}_1(\theta^1)^\dagger \right]^2 \end{aligned} \quad (16)$$

$$\geq \mathbb{E}_{\theta^2} \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^S \text{Tr} [Z_i \text{RX}_1(\theta^2) \rho'_{L-1} \text{RX}_1(\theta^2)^\dagger]^2 \quad (17)$$

$$\geq \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^{2S} \text{Tr} [Z_i \rho'_{L-1}]^2, \quad (18)$$

146 where Eq. (16) is derived by noticing that  $Z_i$  commutes with  $\text{CZ}_1$ . Eqs. (17) and (18) yields from  
 147 Lemma 2. By applying the derivation in Eqs. (16-18) for  $j-1$  times, we obtain

$$\mathbb{E}_{\theta^1, \theta^2, \dots, \theta^{2j-2}} \text{Tr} [Z_i \rho'_L]^2 \geq \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^{2(j-1)S} \text{Tr} [Z_i \rho'_{L+1-j}]^2. \quad (19)$$

148 Next, we deal with parameters  $\theta^{2j-1}$  and  $\theta^{2j}$ . For the case of  $m = 2j-1$ , i.e., the partial derivative  
 149 corresponds to the RY layer, by denoting  $\sigma := \text{RX}_j(\theta^{2j}) \rho_{L-j} \text{RX}_j(\theta^{2j})^\dagger$ , we have

$$\begin{aligned} &\mathbb{E}_{\theta^{2j-1}, \theta^{2j}} \text{Tr} [Z_i \rho'_{L+1-j}]^2 \\ &= \mathbb{E}_{\theta^{2j-1}, \theta^{2j}} \frac{1}{4} \left( \text{Tr} \left[ Z_i \text{CZ}_j \text{RY}_j(\theta_+^{2j-1}) \sigma \text{RY}_j(\theta_+^{2j-1})^\dagger \text{CZ}_j^\dagger \right] \right. \\ &\quad \left. - \text{Tr} \left[ Z_i \text{CZ}_j \text{RY}_j(\theta_-^{2j-1}) \sigma \text{RY}_j(\theta_-^{2j-1})^\dagger \text{CZ}_j^\dagger \right] \right)^2 \\ &= \mathbb{E}_{\theta^{2j-1}, \theta^{2j}} \frac{1}{4} \left( \text{Tr} \left[ Z_i \text{RY}_j(\theta_+^{2j-1}) \sigma \text{RY}_j(\theta_+^{2j-1})^\dagger \right] - \text{Tr} \left[ Z_i \text{RY}_j(\theta_-^{2j-1}) \sigma \text{RY}_j(\theta_-^{2j-1})^\dagger \right] \right)^2 \end{aligned} \quad (20)$$

$$= \mathbb{E}_{\theta^{2j-1}, \theta^{2j}} \left( \frac{\partial}{\partial \theta^{2j-1,n}} \text{Tr} [Z_i \text{RY}_j(\theta^{2j-1}) \sigma \text{RY}_j(\theta^{2j-1})^\dagger] \right)^2 \quad (21)$$

$$\geq (1 - \delta_{i_n,0}) \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^{S-1} \frac{1 - \exp[-2\gamma^2]}{2} \mathbb{E}_{\theta^{2j}} \text{Tr} [Z_i \sigma]^2 \quad (22)$$

$$\geq (1 - \delta_{i_n,0}) \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^{2S-1} \frac{1 - \exp[-2\gamma^2]}{2} \text{Tr} [Z_i \rho_{L-j}]^2, \quad (23)$$

where Eq. (20) is derived by noticing that  $Z_i$  commutes with  $CZ_j$ . Eq. (21) is derived by using the parameter-shift rule. Eqs. (22) and (23) are obtained by using Lemma 2. For the case of  $m = 2j$ , the result in Eq. (23) can be derived similarly. Next, by combining the derivation in Eqs. (13-23), we have

$$\begin{aligned}\mathbb{E}_{\theta} \text{Tr}[Z_i \rho'_L]^2 &\geq (1 - \delta_{i_n 0}) \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^{2LS-1} \frac{1 - \exp[-2\gamma^2]}{2} \text{Tr}[Z_i \rho_0]^2 \\ &= (1 - \delta_{i_n 0}) \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^{2LS-1} \frac{1 - \exp[-2\gamma^2]}{2}\end{aligned}\quad (24)$$

$$\begin{aligned}&= (1 - \delta_{i_n 0}) \left( \frac{1 + \exp[-2\gamma^2]}{2} \right)^{2LS} \frac{1 - \exp[-2\gamma^2]}{1 + \exp[-2\gamma^2]} \\ &\geq (1 - \delta_{i_n 0}) (1 - \gamma^2)^{2LS} \frac{2}{3} \gamma^2,\end{aligned}\quad (25)$$

where Eq. (24) follows from  $\text{Tr}[Z_i|0\rangle\langle 0|] = 1$ . Eq. (25) is obtained by using  $\exp[-x] \geq 1 - x$  and  $\frac{1 - \exp[-x]}{1 + \exp[-x]} \geq \frac{x}{3}$  for  $0 \leq x \leq 2$ . Applying Eq. (24) in Eq. (15) yields

$$\mathbb{E}_{U, \theta} \left( \frac{\partial f}{\partial \theta^{m,n}} \right)^2 \geq \frac{1}{3^S} \sum_i \alpha_i^2 (1 - \delta_{i_n 0}) (1 - \gamma^2)^{2LS} \frac{2}{3} \gamma^2. \quad (26)$$

Moreover, for the case of local observables with  $S = \mathcal{O}(1)$  and  $\alpha_i = \mathcal{O}(1)$ , we have

$$\begin{aligned}\mathbb{E}_{U, \theta} \|\nabla_{\theta} f\|^2 &= \sum_{m=1}^{2L} \sum_{n=1}^N \mathbb{E}_{U, \theta} \left( \frac{\partial f}{\partial \theta^{m,n}} \right)^2 \\ &\geq \sum_{m=1}^{2L} \sum_{n=1}^N \frac{1}{3^S} \sum_i \alpha_i^2 (1 - \delta_{i_n 0}) (1 - \gamma^2)^{2LS} \frac{2}{3} \gamma^2 \\ &\geq \mathcal{O}(1)\end{aligned}\quad (27)$$

by choosing  $\gamma^2 = c/L$ .

□

## D Backbone ResNet-18 Training

### Problem Statement for Anisotropic Hamiltonian

Let  $\{(\mathbf{x}_i, \mathbf{c}_i)\}_{i=1}^N$  be a training dataset, where each  $\mathbf{x}_i \in \mathbb{R}^{C \times H \times W}$  is a multi-channel 2D input image/tensor (in our case,  $C = 5$  for the coordinate channels plus the parameters  $(a, b, c)$ ). Each corresponding ground truth  $\mathbf{c}_i \in \mathbb{R}^{1 \times H \times W}$  is the target intensity map.

We denote the model parameters collectively by  $\theta$ . The model, which we write as a function  $f_{\theta}(\mathbf{x})$ , outputs a predicted intensity map of size  $\mathbb{R}^{1 \times H \times W}$ . Specifically, the network architecture can be expressed as a composition of layers:

$$f_{\theta}(\mathbf{x}) = \text{ConvOut}\left(\text{MHAttn2D}(\text{ResNet18}(\mathbf{x}))\right),$$

where:

- ResNet18Backbone denotes the modified ResNet18 layers (first convolution, no max pooling, and adjusted strides).
- MHAttn2D is the multi-head self-attention module in 2D.
- ConvOut is the final  $1 \times 1$  convolution layer projecting to 1 channel.

### Loss Function

173 We use the Mean Squared Error (MSE) as the training loss. For a single training example  $(\mathbf{x}, \mathbf{c})$ , the  
 174 MSE loss is given by

$$\mathcal{L}(\mathbf{c}, f_{\theta}(\mathbf{x})) = \frac{1}{HW} \sum_{h=1}^H \sum_{w=1}^W \left( y_{h,w} - \hat{c}_{h,w}(\theta) \right)^2 = \frac{1}{HW} \|\mathbf{c} - f_{\theta}(\mathbf{x})\|^2,$$

175 where  $\hat{c}_{h,w}(\theta)$  represents the predicted pixel value at position  $(h, w)$ , and  $c_{h,w}$  is the corresponding  
 176 ground truth pixel value.

177 Given a minibatch of size  $B$  (the default in our script is often  $B = 1$ , but let us write it in general  
 178 form), the total MSE loss is the average over the batch:

$$\mathcal{L}_{\text{batch}}(\theta) = \frac{1}{B} \sum_{i=1}^B \frac{1}{HW} \|\mathbf{c}_i - f_{\theta}(\mathbf{x}_i)\|^2.$$

### 179 Optimization via Gradient Descent

180 We seek to find parameters  $\theta$  that minimize  $\mathcal{L}_{\text{batch}}(\theta)$ . In gradient-based optimization, we iteratively  
 181 update  $\theta$  in the direction of the negative gradient of  $\mathcal{L}_{\text{batch}}$ . If we let  $\eta$  denote the learning rate, then  
 182 the parameter update rule in standard gradient descent is:

$$\theta \leftarrow \theta - \eta \nabla_{\theta} \mathcal{L}_{\text{batch}}(\theta).$$

183 In our code, we use the Adam optimizer, which introduces adaptive learning rates and momentum  
 184 terms. Conceptually, the Adam update can be written as:

$$\begin{aligned} m_t &\leftarrow \beta_1 m_{t-1} + (1 - \beta_1) \nabla_{\theta} \mathcal{L}_{\text{batch}}(\theta), \\ v_t &\leftarrow \beta_2 v_{t-1} + (1 - \beta_2) (\nabla_{\theta} \mathcal{L}_{\text{batch}}(\theta))^2, \\ \hat{m}_t &\leftarrow \frac{m_t}{1 - \beta_1^t}, \\ \hat{v}_t &\leftarrow \frac{v_t}{1 - \beta_2^t}, \\ \theta &\leftarrow \theta - \eta \frac{\hat{m}_t}{\sqrt{\hat{v}_t} + \epsilon}, \end{aligned}$$

185 where  $\beta_1, \beta_2$  are hyperparameters (often 0.9 and 0.999), and  $\epsilon$  is a small constant (e.g.,  $10^{-8}$ ) to  
 186 prevent division by zero. These moment estimates help achieve faster and more stable convergence in  
 187 practice.

### 188 Forward Pass

189 1. **Input Preparation:** Each input  $\mathbf{x}_i$  is a 5-channel tensor of dimension  $(5, H, W)$ . For  
 190 instance:

$$\mathbf{x}_i = (x_{i,\text{coordLayer}}, x_{i,\text{coordCol}}, x_{i,a}, x_{i,b}, x_{i,c}).$$

191 2. **ResNet18 Backbone:** The first convolutional layer, batch norm, and subsequent residual  
 192 blocks produce an intermediate feature map:

$$\mathbf{f}_{\text{res}} = \text{ResNet18Backbone}(\mathbf{x}_i).$$

193 3. **2D Multi-Head Self-Attention:** We apply a 2D multi-head self-attention module to  $\mathbf{f}_{\text{res}}$ .  
 194 This module computes queries  $\mathbf{Q}$ , keys  $\mathbf{K}$ , and values  $\mathbf{V}$  across spatial positions:

$$\mathbf{Q} = W_q * \mathbf{f}_{\text{res}}, \quad \mathbf{K} = W_k * \mathbf{f}_{\text{res}}, \quad \mathbf{V} = W_v * \mathbf{f}_{\text{res}},$$

195 where  $*$  denotes a  $1 \times 1$  convolution, and  $W_q, W_k, W_v$  are the learned parameters. We split  
 196 these into  $H_{\text{heads}}$  heads, each of dimension  $d_k$  per head. The attention weights for each head  
 197  $h$  are given by:

$$\alpha_h(\mathbf{Q}, \mathbf{K}) = \text{Softmax}(\mathbf{Q}_h \mathbf{K}_h^{\top}),$$

198 and the output of the attention for head  $h$  is

$$\mathbf{O}_h = \mathbf{V}_h \alpha_h^{\top}.$$

199 The combined output across all heads is then projected and added to the input feature map  
 200 (via a residual connection multiplied by a learnable scalar  $\gamma$ ):

$$\mathbf{f}_{\text{attn}} = \mathbf{f}_{\text{res}} + \gamma \left[ W_o \left( \text{Concat}_{h=1}^{H_{\text{heads}}} (\mathbf{O}_h) \right) \right].$$



201 **4. Final  $1 \times 1$  Convolution:** We pass  $\mathbf{f}_{\text{attn}}$  through a final convolution to predict a single-channel  
 202 intensity map:

$$\hat{\mathbf{c}}_i = \text{Conv}_{1 \times 1}(\mathbf{f}_{\text{attn}}).$$

203 **5. Upsampling (if needed):** If the ResNet blocks reduce spatial resolution, we may apply  
 204 bilinear upsampling to match the original height  $H$  and width  $W$ :

$$\hat{\mathbf{c}}_i = \text{Upsample}(\hat{\mathbf{c}}_i, (H, W)).$$

## 205 Backward Pass

206 We define the MSE loss for the  $i$ -th example:

$$\mathcal{L}(\theta; \mathbf{x}_i, \mathbf{c}_i) = \frac{1}{HW} \|\mathbf{c}_i - \hat{\mathbf{c}}_i\|^2.$$

207 Then, we compute the gradient with respect to  $\theta$  by applying the chain rule:

$$\nabla_{\theta} \mathcal{L}(\theta; \mathbf{x}_i, \mathbf{c}_i) = \frac{\partial \mathcal{L}}{\partial \hat{\mathbf{c}}_i} \frac{\partial \hat{\mathbf{c}}_i}{\partial \mathbf{f}_{\text{attn}}} \frac{\partial \mathbf{f}_{\text{attn}}}{\partial \mathbf{f}_{\text{res}}} \frac{\partial \mathbf{f}_{\text{res}}}{\partial \theta}.$$

208 In practice, modern deep learning libraries (e.g., PyTorch) automate this via backpropagation,  
 209 computing partial derivatives layer by layer. The final step is an optimizer update (e.g., Adam) based  
 210 on these gradients.

211 The overall training loop can be summarized as follows:

- 212 1. **Initialize model parameters  $\theta$ .**
- 213 2. **For each epoch  $e \in \{1, 2, \dots, E\}$ :**
  - 214 (a) Shuffle dataset  $\{(\mathbf{x}_i, \mathbf{c}_i)\}_{i=1}^N$  and (optionally) form mini-batches of size  $B$ .
  - 215 (b) **For each mini-batch  $\mathcal{B}$ :**
    - 216 i. Perform a forward pass:  $\hat{\mathbf{c}}_i = f_{\theta}(\mathbf{x}_i)$  for each  $(\mathbf{x}_i, \mathbf{c}_i) \in \mathcal{B}$ .
    - 217 ii. Compute the MSE loss over the batch:

$$\mathcal{L}_{\text{batch}}(\theta) = \frac{1}{B} \sum_{i \in \mathcal{B}} \frac{1}{HW} \|\mathbf{c}_i - \hat{\mathbf{c}}_i\|^2.$$

- 218 iii. Backpropagate to obtain  $\nabla_{\theta} \mathcal{L}_{\text{batch}}(\theta)$ .
- 219 iv. Update parameters  $\theta$  using Adam or standard gradient descent.

220 After convergence (or after a chosen number of epochs), we obtain a trained model  $f_{\theta}$  that can be  
 221 used to predict freeze intensity maps for new inputs.

## 222 E TITAN: Complementary to other methods

### 223 E.1 Layer-wise Greedy Gradient Descent (LGGD)

224 Let a hardware-efficient VQE ansatz acting on  $N$  qubits be written as a depth-factorised unitary

$$U(\theta) = \prod_{\ell=1}^L U_{\ell}(\theta^{(\ell)}), \quad U_{\ell} = [\text{CZ} \cdot \text{RX} \cdot \text{RY}]^{\otimes N}, \quad (28)$$

225 where  $\theta = (\theta^{(1)}, \dots, \theta^{(L)})$  and  $\theta^{(\ell)} \in \mathbb{R}^{2N}$  contains the RX/RX angles of layer  $\ell$ . Throughout  
 226 we denote by  $\theta_{1:\ell} = (\theta^{(1)}, \dots, \theta^{(\ell)})$  the *prefix* up to depth  $\ell$ . The VQE objective for a target  
 227 Hamiltonian  $H = \sum_j h_j P_j$  is

$$\mathcal{L}(\theta) = \langle 0 | U(\theta)^{\dagger} H U(\theta) | 0 \rangle, \quad \text{to be minimised.} \quad (29)$$

228 **Greedy depth-growth schedule.** Starting from a shallow prefix of depth  $\ell = 1$  we minimise

$$\mathcal{L}_{\ell}(\theta_{1:\ell}) := \mathcal{L}(\theta_{1:\ell}, \underbrace{\mathbf{0}, \dots, \mathbf{0}}_{L-\ell \text{ frozen layers}}), \quad (30)$$

229 *keeping all yet-to-be-added layers fixed at 0.* After  $T$  gradient steps we **freeze**  $\theta^{(\ell)}$  and expose  $\theta^{(\ell+1)}$ ;  
 230 the process repeats until  $\ell = L$ . The update rule for the active prefix is the usual GD step,

$$\theta_{t+1}^{(\ell)} = \theta_t^{(\ell)} - \eta \nabla_{\theta^{(\ell)}} \mathcal{L}_{\ell}(\theta_t), \quad (31)$$

231 where  $\eta$  is the learning rate.

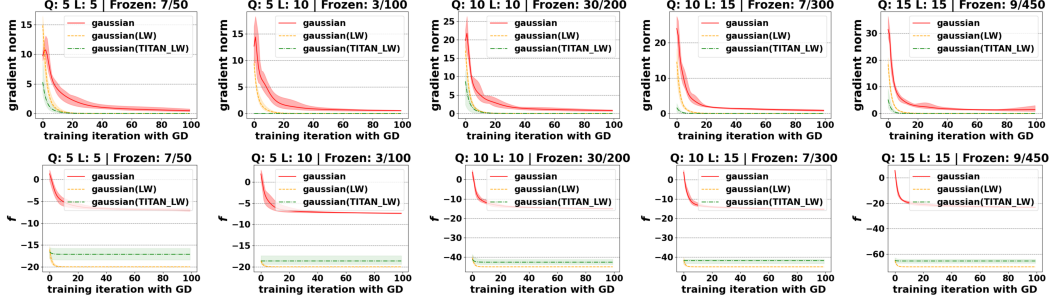


Figure 3: Convergence behavior of TITAN + LAYERWISE (green dotted) versus the *Baseline* (solid red) and a *Layerwise* control (yellow dotted) HEA isotropic Hamiltonian with Gaussian initialization [20]. The **top row** plots the  $\ell_2$ -norm, while the **bottom row** tracks  $f$  (final energy). Shaded envelopes denote  $\pm\sigma$  across **5 independent runs** with threshold ( $\tau = 80$ ).

**Gradient consistency.** Because the expectation in (30) depends on the prefix *only through*  $U_{1:\ell} \equiv \prod_{k=1}^{\ell} U_k$ , one has

$$\nabla_{\theta^{(\ell)}} \mathcal{L} = \nabla_{\theta^{(\ell)}} \mathcal{L}_{\ell} \quad \nabla_{\theta^{(k)}} \mathcal{L}_{\ell} = \mathbf{0}, \quad \forall k > \ell,$$

hence LGGD reproduces the exact gradients of the full objective *restricted to* the current layer set and incurs no bias. The per-iteration cost is therefore  $\mathcal{O}(\ell \text{ poly}(N))$  instead of  $\mathcal{O}(L \text{ poly}(N))$ , yielding a cumulative complexity  $\sum_{\ell=1}^L \mathcal{O}(\ell) = \mathcal{O}(L^2)$  circuit evaluations—quadratically smaller than standard depth- $L$  training.

**Convergence guarantee (sketch).** Assume each layer unitary  $U_{\ell}(\cdot)$  is  $\beta$ -smooth and the objective is lower-bounded by  $\mathcal{L}_{\star}$ . For a fixed depth  $\ell$  and step size  $0 < \eta \leq 1/\beta$ ,

$$\mathcal{L}_{\ell}(\theta_t) - \mathcal{L}_{\star} \leq \frac{\|\nabla \mathcal{L}_{\ell}(\theta_0)\|^2}{2\beta t},$$

mirroring classical GD. Thus each stage achieves  $\varepsilon$ -accuracy in  $T = \mathcal{O}(\beta/\varepsilon)$  steps, and the overall runtime is  $\mathcal{O}(L^2\beta/\varepsilon)$ .

**Discussion.** LGGD based Layerwise (LW) method bears a resemblance to curriculum-style training in deep networks and to the *Disentangled ADAPT-VQE*[8] strategy, but differs in that (i) it does not search for new gates; (ii) its depth growth is deterministic; and (iii) intermediate gradients are exact for the active sub-circuit. As illustrated in Figure 3, the LGGD-based layer-wise (LW) initialization surpasses conventional Gaussian initialization, achieving both faster convergence and a lower final energy. More critically, the TITAN framework interfaces seamlessly with either LGGD-based LW or Gaussian initialization. When coupled with LGGD-based LW, TITAN preserves the same minimum-energy solution quality while *further* accelerating convergence; in addition, the parameter-freezing mechanism embedded in TITAN reduces the measurement overhead, thereby enhancing experimental efficiency without sacrificing accuracy.

## F Experiments

**TITAN Training Settings** For Dataset Construction, all runs sweep the circuit width  $n_{\text{qubits}} \in \{5, \dots, 15\}$  and depth  $L \in \{5, \dots, 15\}$ , train each setting for 100 gradient steps, repeat the experiment 20 times, and print diagnostics every 20 epochs. Learning rates are fixed at  $\eta = 0.01$ . For gradient-descent with APFA, we decay the per-parameter EMA with  $\alpha = 0.7$  and freeze a weight once its EMA stays below  $\tau_f = 0.2 \bar{g}$  for  $N_f = 3$  consecutive steps; re-activation occurs when the EMA exceeds  $\tau_a = 0.4 \tau_f$  for  $N_a = 2$  steps after an initial 20-step warm-up. The Adam variant inherits the same  $\alpha$  and patience lengths but tightens the thresholds to  $\tau_f = 0.5 \bar{g}$  and  $\tau_a = 1.0 \phi_f$ ; its momentum parameters are  $\beta_1 = 0.9$ ,  $\beta_2 = 0.99$  with numerical stabilizer  $\varepsilon = 10^{-8}$ .

For model training, the learning rate is set to be 0.001 and training epochs are 1200, optimizer is Adam.

263 **Isotropic Hamiltonian & Anisotropic Hamiltonian** Train each setting for 100 gradient steps, repeat  
 264 the experiment 20 times, and print diagnostics every 20 epochs. Learning rates are fixed at  $\eta = 0.01$ .  
 265 Optimizer is Gradient Descent (GD).

## 266 F.1 Supplementary experiments and results: Molecule

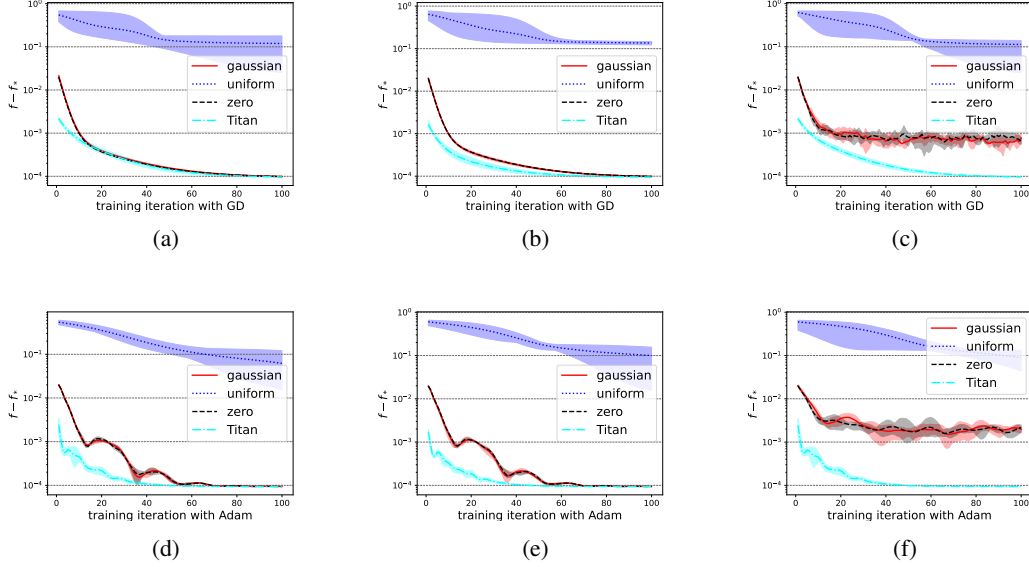


Figure 4: Numerical results of loss  $f$  minus global minimum  $f_*$  of the molecule LiH. The first and second rows show training results with the gradient descent and the Adam optimizer, respectively. The left, the middle, and the right columns show results using accurate gradients, noisy gradients with adaptive-distributed noises, and noisy gradients with constant-distributed noises. The variance of noises in the middle line (Figures 4b and 4e), while the variance of noises in the right line (Figures 4c and 4f) is 0.001. Each line denotes the average of 5 rounds of optimizations.

267 For an active space of  $n_e$  electrons in  $n_0$  spin-orbitals, the second-quantized encoding employs  
 268  $N = n_0$  qubits, each representing a single spin-orbital occupation. The mean-field reference  
 269 is  $|\phi_{\text{HF}}\rangle = \bigotimes_{j=1}^{n_e} |1\rangle_j \otimes \bigotimes_{j=n_e+1}^{n_0} |0\rangle_j$ , where  $|1\rangle$  ( $|0\rangle$ ) denotes an occupied (vacant) spin-  
 270 orbital. Electron correlation is introduced via a sequence of Givens rotations,  $V_{\text{Givens}}(\theta) =$   
 271  $\prod_{l=1}^L R_{\text{Givens}}^{(l)}(\theta_l)$ , with  $L$  two- or four-qubit gates; the value of  $L$  is determined by the chosen orbital  
 272 mapping and basis. The molecular electronic Hamiltonian in second quantization reads

$$H = \sum_{pq} h_{pq} a_p^\dagger a_q + \frac{1}{2} \sum_{pqrs} h_{pqrs} a_p^\dagger a_q^\dagger a_r a_s, \quad (32)$$

273 which, after a Jordan–Wigner or Bravyi–Kitaev transform, becomes a sum of Pauli strings on  $N$   
 274 qubits. Ground-state energy with respect to the parameter vector  $\theta$  is approximated by minimizing

$$f(\theta) = \langle \phi_{\text{HF}} | V_{\text{Givens}}^\dagger(\theta) H V_{\text{Givens}}(\theta) | \phi_{\text{HF}} \rangle. \quad (33)$$

275 **Active Electrons and Spin Orbitals** For a molecule with  $n_e$  active electrons and  $n_0$  spin orbitals.  
 276 The dimension of the corresponding Hilbert space on a quantum computer is determined by  $n_0$ ,  
 277 leading to  $N = n_0$  qubits for representing the molecular wavefunction in the second-quantized form.

278 **Hartree–Fock Reference State** A common starting point for a variational algorithm is the Hartree–  
 279 Fock (HF) reference state,

$$|\phi_{\text{HF}}\rangle = \underbrace{|1\rangle \otimes |1\rangle \otimes \cdots \otimes |1\rangle}_{n_e \text{ times}} \otimes \underbrace{|0\rangle \otimes |0\rangle \otimes \cdots \otimes |0\rangle}_{n_0 - n_e \text{ times}}. \quad (34)$$

280 This state encodes the mean-field approximation in which the  $n_e$  active electrons occupy the lowest-  
 281 energy orbitals, and the remaining  $n_0 - n_e$  orbitals are left unoccupied.

**Parameterization via Givens Rotations** To capture electron correlation effects beyond the HF approximation, we apply a parameterized quantum circuit composed of Givens rotations. Denote the unitary operator as  $V_{\text{Givens}}(\theta) = \prod_{i=1}^L R_{\text{Givens}}^{(i)}(\theta_i)$ , where each  $R_{\text{Givens}}^{(i)}(\theta_i)$  is either a 2-qubit or 4-qubit Givens rotation gate, parameterized by an angle  $\theta_i$ . The total number of such gates,  $L$ , depends on the specific molecule (e.g., the basis set, chosen orbital mapping, etc.).

**Molecular Hamiltonian** The second-quantized electronic structure Hamiltonian for a given molecule can be written generally as

$$H_{\text{mol}} = \sum_{p,q} h_{pq} a_p^\dagger a_q + \frac{1}{2} \sum_{p,q,r,s} h_{pqrs} a_p^\dagger a_q^\dagger a_r a_s, \quad (35)$$

where  $a_p^\dagger$  ( $a_p$ ) are the fermionic creation (annihilation) operators, and  $h_{pq}$ ,  $h_{pqrs}$  are the one- and two-electron integrals, respectively. After performing a Jordan-Wigner or Bravyi-Kitaev transformation,  $H_{\text{mol}}$  can be expressed as a sum of Pauli operators on  $N$  qubits.

**Variational Loss Function** Our goal is to minimize the expectation value of  $H_{\text{mol}}$  with respect to the parameterized state. Formally, we define the loss function

$$f(\theta) = \text{Tr} \left[ H_{\text{mol}} V_{\text{Givens}}(\theta) |\phi_{\text{HF}}\rangle \langle \phi_{\text{HF}}| V_{\text{Givens}}(\theta)^\dagger \right]. \quad (36)$$

By varying the set of parameters  $\{\theta_i\}$  and minimizing  $f(\theta)$ , one effectively searches for the lowest eigenvalue of  $H_{\text{mol}}$ , which corresponds to the ground-state energy of the molecule.

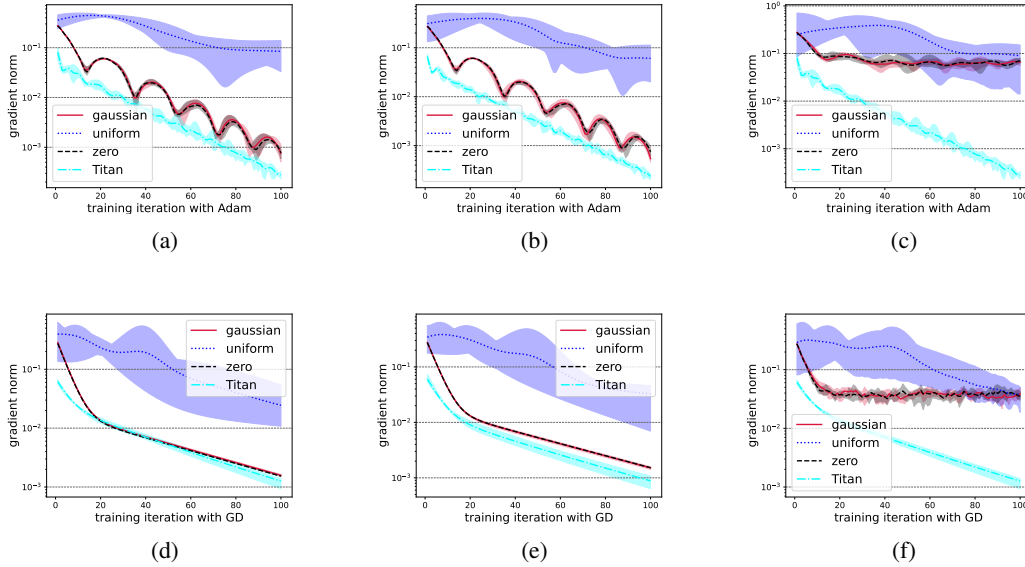
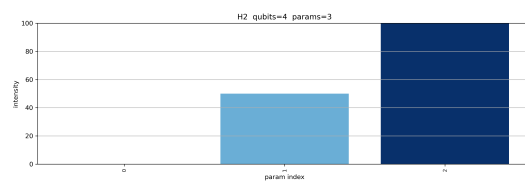


Figure 5: Numerical results of finding the ground energy of the molecule LiH. The first and second rows show training results with the gradient descent and the Adam optimizer, respectively. The left, the middle, and the right columns show results using accurate gradients, noisy gradients with adaptive-distributed noises, and noisy gradients with constant-distributed noises. The variance of noises in the middle line (Figures 5b and 5e), while the variance of noises in the right line (Figures 5c and 5f) is 0.001. Each line denotes the average of 5 rounds of optimizations.

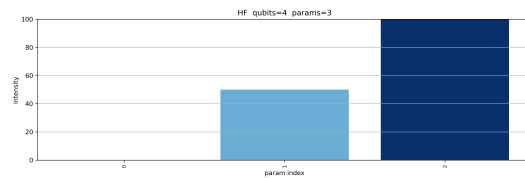
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Table 1: Representative Molecules

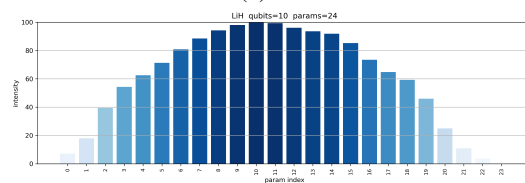
Molecule	$N_e$	$N_{\text{orb}}$	Qubits ( $N$ )
H <sub>2</sub>	2	2	4
HF	2	5	4
LiH	2	5	10
BeH <sub>2</sub>	4	6	12
H <sub>2</sub> O	6	7	10
N <sub>2</sub>	6	6	12
CO	6	7	12



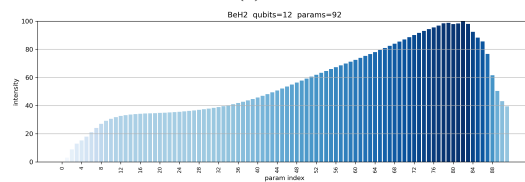
(a) H2



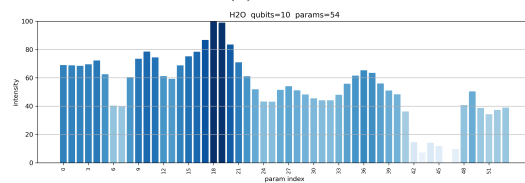
(b) HF



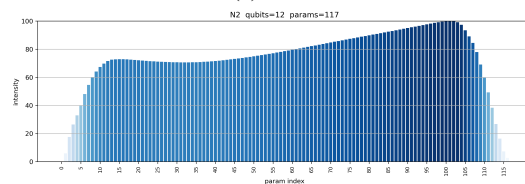
(c) LiH



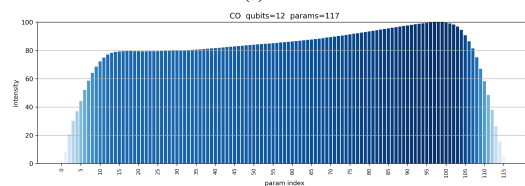
(d) BeH2



(e) H2O



(f) N2



(g) CO

Figure 6: Titan Predicted intensity of different molecules.

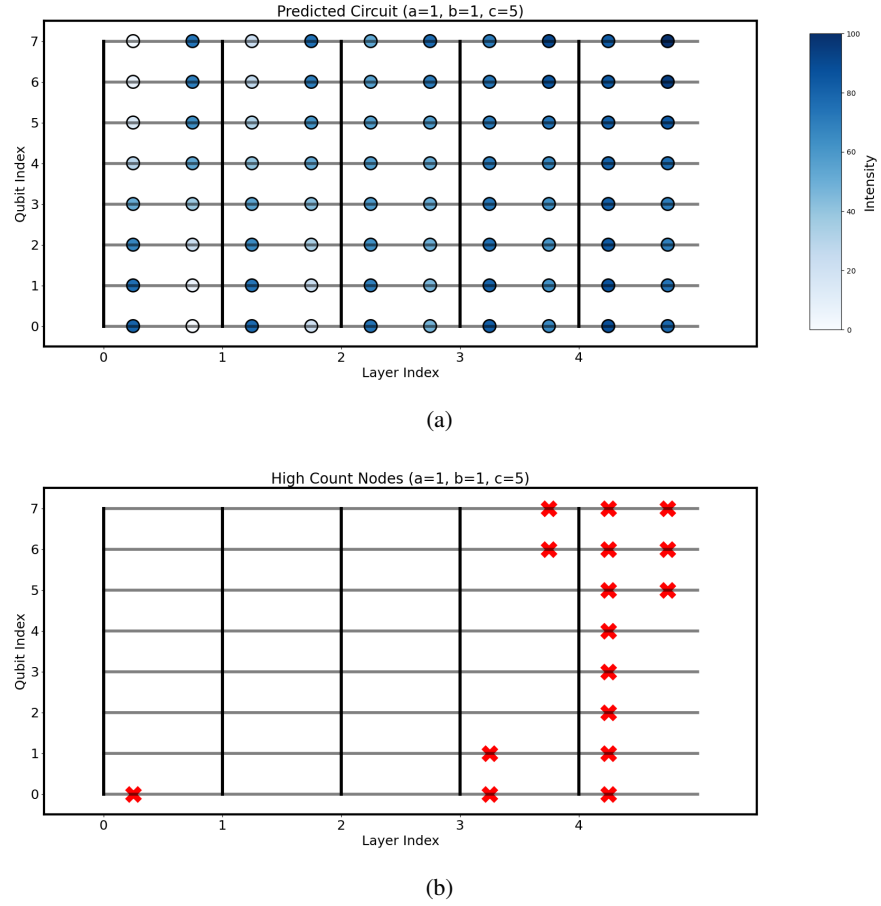


Figure 7: (a) TITAN predicted intensity. (b) TITAN selected frozen gates (**16/80**) (20%). Qubits 8, Layers 5, Threshold:  $\tau = 80$ . HEA-based anisotropic Hamiltonian solver:  $1 \times XX + 1 \times YY + 5 \times ZZ$ .

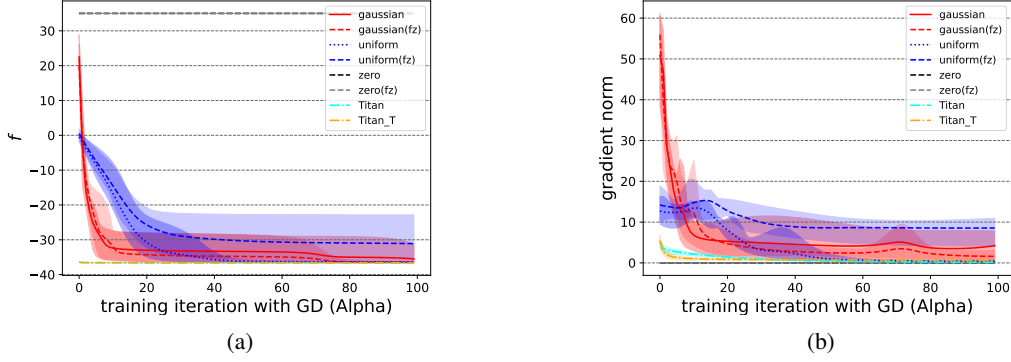


Figure 8: (a) Loss GD Noiseless. (b) Gradient Norm GD Noiseless. Qubits 8, Layers 5, Threshold:  $\tau = 80$ . Selected Frozen Gates (**16/80**) (**20%**). HEA-based anisotropic Hamiltonian solver:  $1 \times XX + 1 \times YY + 5 \times ZZ$ . TITAN: Enhanced Gaussian. TITAN\_T: APFA Only without freezing.

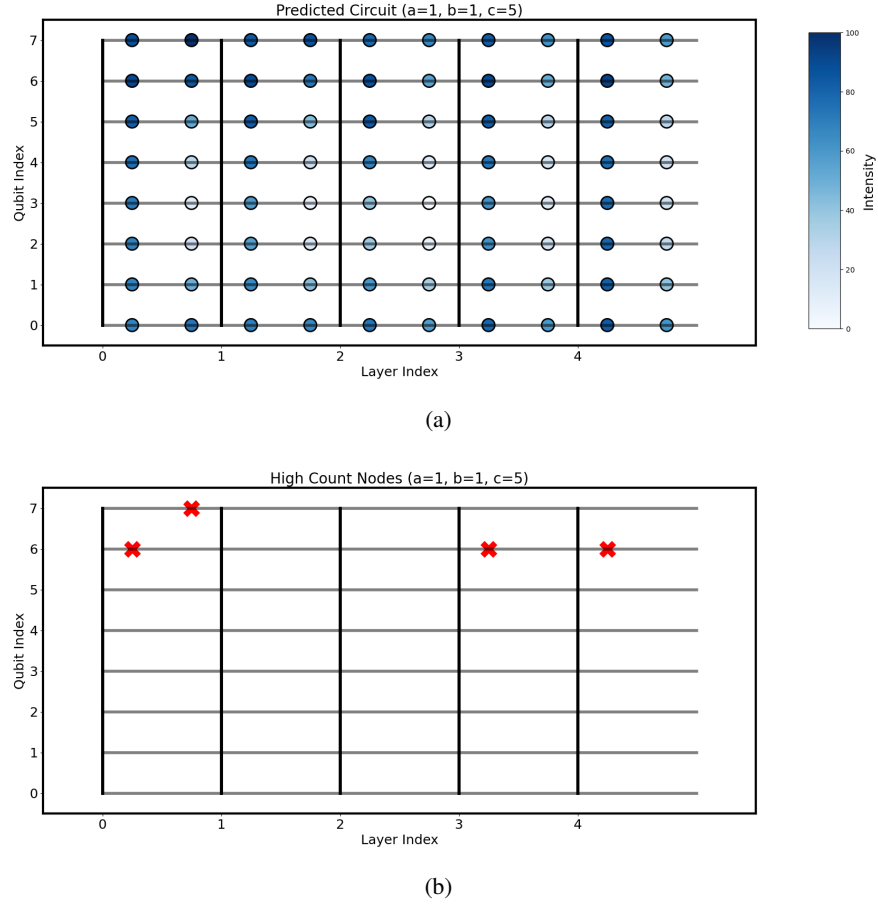


Figure 9: (a) TITAN predicted intensity. (b) TITAN selected Frozen Gates (**4/80**) (**5%**). Qubits 8, Layers 5, Threshold: 90. HEA-based anisotropic Hamiltonian solver:  $1 \times XX + 1 \times YY + 5 \times ZZ$ .



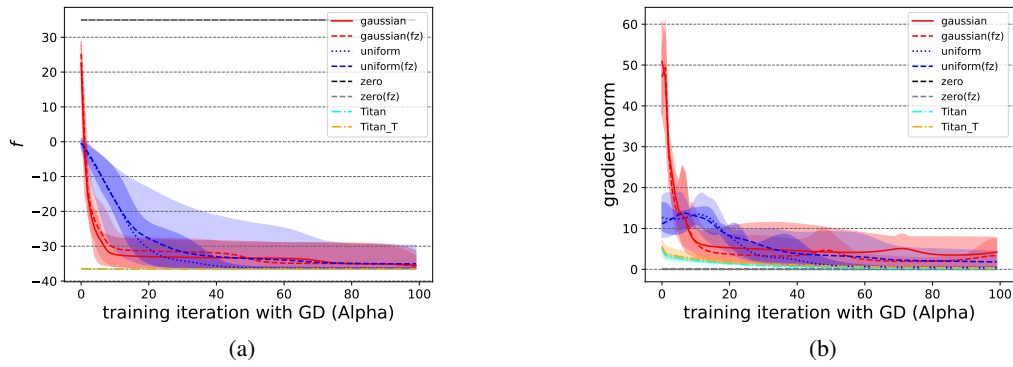


Figure 10: (a) Loss GD Noiseless. (b) Gradient Norm GD Noiseless. Qubits 8, Layers 5, Threshold: 90. Selected Frozen Gates (**4/80**) (5%). HEA-based anisotropic Hamiltonian solver:  $1 \times XX + 1 \times YY + 5 \times ZZ$ . TITAN: Enhanced Gaussian. TITAN\_T: APFA Only without freezing.

Table 2: Frozen performance comparison between TITAN and Random Freezing (ansätze: HEA, Optimizer: GD). Frozen-parameter counts that exceed 10% are highlighted in blue; entries with  $\Delta E \leq 0$  are highlighted in green.

	5	6	7	8	9	10	11	12	13	14	15
<b>Proposed TITAN: Final Energy Comparison (<math>\Delta E = E_{\text{Titan Gauss}} - E_{\text{Baseline Gauss}}</math>)</b>											
<b>5</b>	0.161	0.145	0.152	0.079	0.374	0.198	<b>-0.092</b>	0.315	0.465	0.203	0.260
<b>6</b>	<b>-0.084</b>	0.109	0.100	0.301	<b>-0.014</b>	0.147	0.216	0.329	0.096	0.922	0.222
<b>7</b>	<b>-0.222</b>	<b>-0.025</b>	0.268	0.235	0.202	0.264	<b>-0.002</b>	<b>-0.008</b>	0.254	0.088	0.324
<b>8</b>	0.108	<b>-0.022</b>	0.123	0.066	0.102	0.089	<b>-0.029</b>	0.147	0.152	0.035	0.215
<b>9</b>	0.019	<b>-0.024</b>	<b>-0.114</b>	<b>-0.030</b>	0.017	<b>-0.120</b>	0.220	<b>-0.114</b>	<b>-0.177</b>	-0.025	<b>-0.063</b>
<b>10</b>	<b>-0.084</b>	0.027	0.029	0.002	0.055	0.104	<b>-0.033</b>	0.131	0.115	0.113	0.289
<b>Frozen Parameters (threshold <math>\tau = 80</math>)</b>											
<b>5</b>	<b>7(14.0%)</b>	5(8.3%)	<b>9(12.9%)</b>	5(6.3%)	<b>15(16.7%)</b>	7(7%)	<b>12(10.9%)</b>	<b>13(10.8%)</b>	10(7.7%)	<b>20(14.3%)</b>	<b>16(10.7%)</b>
<b>6</b>	4(6.7%)	4(5.6%)	<b>15(17.9%)</b>	8(8.3%)	10(9.3%)	8(6.7%)	12(9.1%)	<b>25(17.3%)</b>	<b>25(16.0%)</b>	<b>31(18.5%)</b>	<b>27(15.0%)</b>
<b>7</b>	5(7.1%)	5(6.0%)	4(4.1%)	10(8.9%)	9(7.1%)	2(1.4%)	6(3.9%)	10(6.0%)	7(3.9%)	5(2.6%)	19(9.1%)
<b>8</b>	4(5.0%)	9(9.4%)	4(3.6%)	<b>14(10.9%)</b>	7(4.9%)	11(6.9%)	<b>22(12.5%)</b>	<b>21(10.9%)</b>	8(3.9%)	6(2.7%)	8(3.3%)
<b>9</b>	2(2.2%)	1(0.9%)	10(7.9%)	2(1.4%)	8(4.9%)	9(5.0%)	<b>21(10.6%)</b>	10(4.6%)	11(4.7%)	8(3.2%)	10(3.7%)
<b>10</b>	3(3%)	12(10.0%)	7(5.0%)	<b>18(11.3%)</b>	11(6.1%)	<b>30(15.0%)</b>	8(3.6%)	4(1.7%)	6(2.3%)	24(8.6%)	18(6.0%)
<b>Final Energy Comparison (<math>\Delta E = E_{\text{Random Gauss}} - E_{\text{Baseline Gauss}}</math>)</b>											
<b>5</b>	0.184	0.153	0.099	0.042	0.118	0.122	0.026	0.089	0.380	0.615	0.466
<b>6</b>	0.106	0.160	0.107	0.139	0.103	0.090	0.126	0.094	0.147	0.460	0.216
<b>7</b>	0.118	0.004	0.047	0.083	0.027	0.025	0.159	0.029	0.163	0.255	0.375
<b>8</b>	0.041	0.123	0.022	0.066	0.026	0.018	0.047	0.075	0.222	0.025	0.173
<b>9</b>	0.052	0.005	0.095	0.072	0.010	<b>-0.069</b>	0.049	0.051	0.223	0.042	0.037
<b>10</b>	0.113	0.022	0.003	0.032	0.015	0.015	<b>-0.068</b>	0.041	<b>-0.097</b>	0.046	0.268

Table 3: Frozen performance comparison between TITAN and Random Freezing (ansätze: HEA, Optimizer: GD). Frozen parameters exceeding between 10% and 30% are colored with blue. The value in each cell is written as  $\Delta E^{(|\Delta E/E_{\text{baseline}}| \%)}$ ; cells where  $\Delta E \leq 0$  are shaded with green of Titan and red of random freezing.

Layers \ Qubit	Qubit: 5	Qubit: 6	Qubit: 7	Qubit: 8	Qubit: 9	Qubit: 10	Qubit: 11	Qubit: 12	Qubit: 13	Qubit: 14	Qubit: 15
<b>Proposed TITAN: Final Energy Comparison: <math>\Delta E = E_{\text{Titan Gaussian}} - E_{\text{Baseline Gaussian}}</math></b>											
Layers: 5	0.161(2.3%)	0.145(1.8%)	0.152(1.5%)	0.079(0.7%)	0.374(2.9%)	0.198(1.4%)	<span style="background-color: #e0ffe0;">-0.092(0.6%)</span>	0.315(1.8%)	0.465(2.5%)	0.203(1.0%)	0.260(1.2%)
Layers: 6	<span style="background-color: #e0ffe0;">-0.084(0.2%)</span>	0.109(1.3%)	0.100(1.0%)	0.301(2.6%)	<span style="background-color: #e0ffe0;">-0.014(0.1%)</span>	0.147(1.0%)	<span style="background-color: #e0ffe0;">0.216(1.3%)</span>	0.329(1.9%)	0.096(0.5%)	0.922(4.5%)	0.222(1.0%)
Layers: 7	<span style="background-color: #e0ffe0;">-0.222(3.1%)</span>	<span style="background-color: #e0ffe0;">-0.025(0.3%)</span>	0.268(2.6%)	0.235(2.0%)	0.202(1.5%)	0.264(1.8%)	<span style="background-color: #e0ffe0;">-0.002(0.0%)</span>	<span style="background-color: #e0ffe0;">-0.008(0.0%)</span>	0.254(1.3%)	0.088(0.4%)	0.324(1.4%)
Layers: 8	0.108(1.5%)	<span style="background-color: #e0ffe0;">-0.022(0.3%)</span>	0.123(1.2%)	0.066(0.6%)	0.102(0.8%)	0.089(0.6%)	<span style="background-color: #e0ffe0;">-0.029(0.2%)</span>	0.147(0.8%)	0.152(0.8%)	0.035(0.2%)	0.215(1.0%)
Layers: 9	0.019(0.3%)	<span style="background-color: #e0ffe0;">-0.024(0.3%)</span>	<span style="background-color: #e0ffe0;">-0.114(1.1%)</span>	<span style="background-color: #e0ffe0;">-0.030(0.3%)</span>	0.017(0.1%)	<span style="background-color: #e0ffe0;">-0.120(0.8%)</span>	<span style="background-color: #e0ffe0;">0.220(1.3%)</span>	<span style="background-color: #e0ffe0;">-0.114(0.6%)</span>	<span style="background-color: #e0ffe0;">-0.177(0.9%)</span>	<span style="background-color: #e0ffe0;">-0.025(0.1%)</span>	<span style="background-color: #e0ffe0;">-0.063(0.3%)</span>
Layers: 10	<span style="background-color: #e0ffe0;">-0.084(1.1%)</span>	0.027(0.3%)	0.029(0.3%)	0.002(0.0%)	0.055(0.4%)	0.104(0.7%)	<span style="background-color: #e0ffe0;">-0.033(0.2%)</span>	0.131(0.7%)	0.115(0.6%)	0.113(0.5%)	0.289(1.3%)
Layers: 11	0.022(0.3%)	<span style="background-color: #e0ffe0;">-0.002(0.0%)</span>	0.118(1.1%)	<span style="background-color: #e0ffe0;">-0.010(0.1%)</span>	<span style="background-color: #e0ffe0;">-0.169(1.3%)</span>	0.103(0.7%)	0.050(0.3%)	0.123(0.7%)	0.206(1.0%)	0.096(0.4%)	0.095(0.4%)
Layers: 12	0.022(0.3%)	<span style="background-color: #e0ffe0;">-0.001(0.0%)</span>	<span style="background-color: #e0ffe0;">-0.095(0.9%)</span>	0.052(0.4%)	0.075(0.6%)	0.148(1.0%)	0.298(1.8%)	<span style="background-color: #e0ffe0;">-0.088(0.5%)</span>	<span style="background-color: #e0ffe0;">-0.023(0.1%)</span>	<span style="background-color: #e0ffe0;">-0.206(1.0%)</span>	0.083(0.4%)
Layers: 13	0.033(0.4%)	0.039(0.4%)	0.262(2.4%)	<span style="background-color: #e0ffe0;">-0.018(0.2%)</span>	<span style="background-color: #e0ffe0;">-0.157(1.2%)</span>	0.053(0.3%)	<span style="background-color: #e0ffe0;">-0.073(0.4%)</span>	0.037(0.2%)	<span style="background-color: #e0ffe0;">-0.016(0.1%)</span>	<span style="background-color: #e0ffe0;">-0.227(1.1%)</span>	<span style="background-color: #e0ffe0;">-0.13(0.6%)</span>
Layers: 14	<span style="background-color: #e0ffe0;">-0.034(0.5%)</span>	0.018(0.2%)	0.212(2.0%)	<span style="background-color: #e0ffe0;">-0.121(1.0%)</span>	0.008(0.1%)	<span style="background-color: #e0ffe0;">-0.001(0.0%)</span>	0.093(0.6%)	<span style="background-color: #e0ffe0;">-0.215(1.2%)</span>	0.186(0.9%)	0.042(0.2%)	0.201(0.9%)
Layers: 15	<span style="background-color: #e0ffe0;">-0.000(0.0%)</span>	0.076(0.9%)	<span style="background-color: #e0ffe0;">-0.066(0.6%)</span>	0.002(0.0%)	<span style="background-color: #e0ffe0;">-0.154(1.1%)</span>	0.050(0.3%)	<span style="background-color: #e0ffe0;">-0.141(0.8%)</span>	0.094(0.5%)	<span style="background-color: #e0ffe0;">-0.061(0.3%)</span>	<span style="background-color: #e0ffe0;">-0.005(0.0%)</span>	0.033(0.1%)
<b>Frozen Parameters: Threshold: <math>\tau = 80</math></b>											
Layers: 5	<span style="background-color: #e0f0ff;">7(14.0%)</span>	<span style="background-color: #e0f0ff;">5(8.3%)</span>	<span style="background-color: #e0f0ff;">9(12.9%)</span>	<span style="background-color: #e0f0ff;">5(6.3%)</span>	<span style="background-color: #e0f0ff;">15(16.7%)</span>	<span style="background-color: #e0f0ff;">7(7%)</span>	<span style="background-color: #e0f0ff;">12(10.9%)</span>	<span style="background-color: #e0f0ff;">13(10.8%)</span>	<span style="background-color: #e0f0ff;">10(7.7%)</span>	<span style="background-color: #e0f0ff;">20(14.3%)</span>	<span style="background-color: #e0f0ff;">16(10.7%)</span>
Layers: 6	<span style="background-color: #e0f0ff;">4(6.7%)</span>	<span style="background-color: #e0f0ff;">4(5.6%)</span>	<span style="background-color: #e0f0ff;">15(17.9%)</span>	<span style="background-color: #e0f0ff;">8(8.3%)</span>	<span style="background-color: #e0f0ff;">10(9.3%)</span>	<span style="background-color: #e0f0ff;">8(6.7%)</span>	<span style="background-color: #e0f0ff;">12(9.1%)</span>	<span style="background-color: #e0f0ff;">25(17.3%)</span>	<span style="background-color: #e0f0ff;">25(16.0%)</span>	<span style="background-color: #e0f0ff;">31(18.5%)</span>	<span style="background-color: #e0f0ff;">27(15.0%)</span>
Layers: 7	<span style="background-color: #e0f0ff;">5(7.1%)</span>	<span style="background-color: #e0f0ff;">5(6.0%)</span>	<span style="background-color: #e0f0ff;">4(4.1%)</span>	<span style="background-color: #e0f0ff;">10(8.9%)</span>	<span style="background-color: #e0f0ff;">9(7.1%)</span>	<span style="background-color: #e0f0ff;">2(1.4%)</span>	<span style="background-color: #e0f0ff;">6(3.9%)</span>	<span style="background-color: #e0f0ff;">10(6.0%)</span>	<span style="background-color: #e0f0ff;">7(3.9%)</span>	<span style="background-color: #e0f0ff;">5(2.6%)</span>	<span style="background-color: #e0f0ff;">19(9.1%)</span>
Layers: 8	<span style="background-color: #e0f0ff;">4(5.0%)</span>	<span style="background-color: #e0f0ff;">9(9.4%)</span>	<span style="background-color: #e0f0ff;">4(3.6%)</span>	<span style="background-color: #e0f0ff;">14(10.9%)</span>	<span style="background-color: #e0f0ff;">7(4.9%)</span>	<span style="background-color: #e0f0ff;">11(6.9%)</span>	<span style="background-color: #e0f0ff;">22(12.5%)</span>	<span style="background-color: #e0f0ff;">21(10.9%)</span>	<span style="background-color: #e0f0ff;">8(3.9%)</span>	<span style="background-color: #e0f0ff;">6(2.7%)</span>	<span style="background-color: #e0f0ff;">8(3.3%)</span>
Layers: 9	<span style="background-color: #e0f0ff;">2(2.2%)</span>	<span style="background-color: #e0f0ff;">1(0.9%)</span>	<span style="background-color: #e0f0ff;">10(7.9%)</span>	<span style="background-color: #e0f0ff;">2(1.4%)</span>	<span style="background-color: #e0f0ff;">8(4.9%)</span>	<span style="background-color: #e0f0ff;">9(5.0%)</span>	<span style="background-color: #e0f0ff;">21(10.6%)</span>	<span style="background-color: #e0f0ff;">10(4.6%)</span>	<span style="background-color: #e0f0ff;">11(4.7%)</span>	<span style="background-color: #e0f0ff;">8(3.2%)</span>	<span style="background-color: #e0f0ff;">10(3.7%)</span>
Layers: 10	<span style="background-color: #e0f0ff;">3(3%)</span>	<span style="background-color: #e0f0ff;">12(10.0%)</span>	<span style="background-color: #e0f0ff;">7(5.0%)</span>	<span style="background-color: #e0f0ff;">18(11.3%)</span>	<span style="background-color: #e0f0ff;">11(6.1%)</span>	<span style="background-color: #e0f0ff;">30(15.0%)</span>	<span style="background-color: #e0f0ff;">8(3.6%)</span>	<span style="background-color: #e0f0ff;">4(1.7%)</span>	<span style="background-color: #e0f0ff;">6(2.3%)</span>	<span style="background-color: #e0f0ff;">24(8.6%)</span>	<span style="background-color: #e0f0ff;">18(6.0%)</span>
Layers: 11	<span style="background-color: #e0f0ff;">29(26.4%)</span>	<span style="background-color: #e0f0ff;">11(8.3%)</span>	<span style="background-color: #e0f0ff;">14(9.1%)</span>	<span style="background-color: #e0f0ff;">4(2.3%)</span>	<span style="background-color: #e0f0ff;">2(1.0%)</span>	<span style="background-color: #e0f0ff;">11(5.0%)</span>	<span style="background-color: #e0f0ff;">5(2.1%)</span>	<span style="background-color: #e0f0ff;">5(1.9%)</span>	<span style="background-color: #e0f0ff;">5(1.8%)</span>	<span style="background-color: #e0f0ff;">17(5.5%)</span>	<span style="background-color: #e0f0ff;">18(5.5%)</span>
Layers: 12	<span style="background-color: #e0f0ff;">13(10.8%)</span>	<span style="background-color: #e0f0ff;">11(7.6%)</span>	<span style="background-color: #e0f0ff;">6(3.6%)</span>	<span style="background-color: #e0f0ff;">21(10.9%)</span>	<span style="background-color: #e0f0ff;">26(12.0%)</span>	<span style="background-color: #e0f0ff;">4(1.7%)</span>	<span style="background-color: #e0f0ff;">12(4.6%)</span>	<span style="background-color: #e0f0ff;">13(4.5%)</span>	<span style="background-color: #e0f0ff;">10(3.2%)</span>	<span style="background-color: #e0f0ff;">8(2.4%)</span>	<span style="background-color: #e0f0ff;">4(1.1%)</span>
Layers: 13	<span style="background-color: #e0f0ff;">8(6.2%)</span>	<span style="background-color: #e0f0ff;">13(8.3%)</span>	<span style="background-color: #e0f0ff;">21(11.5%)</span>	<span style="background-color: #e0f0ff;">13(6.3%)</span>	<span style="background-color: #e0f0ff;">14(6.0%)</span>	<span style="background-color: #e0f0ff;">8(3.1%)</span>	<span style="background-color: #e0f0ff;">23(8.0%)</span>	<span style="background-color: #e0f0ff;">15(4.8%)</span>	<span style="background-color: #e0f0ff;">20(5.9%)</span>	<span style="background-color: #e0f0ff;">32(8.8%)</span>	<span style="background-color: #e0f0ff;">12(3.1%)</span>
Layers: 14	<span style="background-color: #e0f0ff;">4(2.9%)</span>	<span style="background-color: #e0f0ff;">7(4.2%)</span>	<span style="background-color: #e0f0ff;">22(11.2%)</span>	<span style="background-color: #e0f0ff;">14(6.3%)</span>	<span style="background-color: #e0f0ff;">4(1.6%)</span>	<span style="background-color: #e0f0ff;">12(4.3%)</span>	<span style="background-color: #e0f0ff;">41(13.3%)</span>	<span style="background-color: #e0f0ff;">13(3.9%)</span>	<span style="background-color: #e0f0ff;">9(2.5%)</span>	<span style="background-color: #e0f0ff;">15(3.8%)</span>	<span style="background-color: #e0f0ff;">24(5.7%)</span>
Layers: 15	<span style="background-color: #e0f0ff;">15(10.0%)</span>	<span style="background-color: #e0f0ff;">15(8.3%)</span>	<span style="background-color: #e0f0ff;">15(7.1%)</span>	<span style="background-color: #e0f0ff;">6(2.5%)</span>	<span style="background-color: #e0f0ff;">15(5.6%)</span>	<span style="background-color: #e0f0ff;">7(2.3%)</span>	<span style="background-color: #e0f0ff;">13(3.9%)</span>	<span style="background-color: #e0f0ff;">8(2.2%)</span>	<span style="background-color: #e0f0ff;">9(2.3%)</span>	<span style="background-color: #e0f0ff;">30(7.1%)</span>	<span style="background-color: #e0f0ff;">9(2.0%)</span>
<b>Final Energy Comparison: <math>\Delta E = E_{\text{Random Gaussian}} - E_{\text{Baseline Gaussian}}</math></b>											
Layers: 5	0.184(2.6%)	0.153(1.9%)	0.099(1.0%)	0.042(0.4%)	0.118(1.0%)	0.122(0.9%)	0.026(0.3%)	0.089(0.5%)	0.380(2.0%)	0.615(3.0%)	0.466(2.1%)
Layers: 6	0.106(1.5%)	0.160(1.9%)	0.107(1.0%)	0.139(1.2%)	0.103(0.7%)	0.090(0.6%)	0.126(0.7%)	0.094(0.5%)	0.147(0.7%)	0.460(2.2%)	0.216(1.0%)
Layers: 7	0.118(1.7%)	0.004(0.0%)	0.047(0.5%)	0.083(0.8%)	0.027(0.2%)	0.025(0.2%)	0.156(1.5%)	0.029(0.2%)	0.163(0.8%)	0.255(1.2%)	0.375(1.7%)
Layers: 8	0.041(0.6%)	0.123(1.4%)	0.022(0.2%)	0.066(0.6%)	0.026(0.2%)	0.018(0.1%)	0.047(0.3%)	0.075(0.4%)	0.222(1.1%)	0.025(0.1%)	0.173(0.8%)
Layers: 9	0.052(0.7%)	0.005(0.1%)	0.095(1.0%)	0.005(0.0%)	0.072(0.7%)	0.010(0.1%)	<span style="background-color: #ffe0e0;">-0.069(0.5%)</span>	0.049(0.3%)	0.051(0.3%)	0.223(1.1%)	0.042(0.2%)
Layers: 10	0.113(1.5%)	0.022(0.3%)	0.003(0.0%)	0.032(0.3%)	0.015(0.1%)	0.015(0.1%)	<span style="background-color: #ffe0e0;">-0.068(0.4%)</span>	0.041(0.2%)	<span style="background-color: #ffe0e0;">-0.097(0.5%)</span>	0.046(0.2%)	0.268(1.2%)
Layers: 11	0.237(3.2%)	<span style="background-color: #ffe0e0;">-0.064(0.7%)</span>	0.050(0.5%)	0.029(0.3%)	<span style="background-color: #ffe0e0;">-0.175(1.4%)</span>	0.078(0.5%)	0.053(0.4%)	0.152(0.9%)	0.155(0.8%)	0.221(1.0%)	0.006(0.0%)
Layers: 12	0.070(0.9%)	0.080(0.9%)	<span style="background-color: #ffe0e0;">-0.096(1.0%)</span>	0.004(0.0%)	0.010(0.1%)	0.033(0.2%)	0.140(0.8%)	0.099(0.6%)	<span style="background-color: #ffe0e0;">-0.215(1.0%)</span>	0.237(1.0%)	0.237(1.0%)
Layers: 13	0.010(0.1%)	0.030(0.4%)	0.048(0.5%)	0.022(0.2%)	<span style="background-color: #ffe0e0;">-0.112(0.9%)</span>	0.043(0.3%)	<span style="background-color: #ffe0e0;">-0.071(0.4%)</span>	0.055(0.3%)	0.183(0.9%)	<span style="background-color: #ffe0e0;">-0.194(0.8%)</span>	0.144(0.6%)
Layers: 14	<span style="background-color: #ffe0e0;">-0.045(0.6%)</span>	<span style="background-color: #ffe0e0;">-0.058(0.7%)</span>	0.027(0.3%)	<span style="background-color: #ffe0e0;">-0.021(0.2%)</span>	0.003(0.0%)	0.007(0.0%)	0.044(0.3%)	<span style="background-color: #ffe0e0;">-0.110(0.6%)</span>	<span style="background-color: #ffe0e0;">-0.134(0.6%)</span>	0.081(0.4%)	0.228(1.0%)
Layers: 15	0.026(0.3%)	0.080(1.0%)	<span style="background-color: #ffe0e0;">-0.150(1.5%)</span>	0.029(0.2%)	<span style="background-color: #ffe0e0;">-0.184(1.3%)</span>	0.047(0.3%)	<span style="background-color: #ffe0e0;">-0.098(0.6%)</span>	0.254(1.4%)	0.008(0.0%)	0.089(0.4%)	0.038(0.2%)

Qubit	Qubit: 5	Qubit: 6	Qubit: 7	Qubit: 8	Qubit: 9	Qubit: 10	Qubit: 11	Qubit: 12	Qubit: 13	Qubit: 14	Qubit: 15
Layers											
Threshold:70											
Layers: 5	13 <sup>26.0%</sup>	10 <sup>16.7%</sup>	17 <sup>24.3%</sup>	27 <sup>33.8%</sup>	30 <sup>33.3%</sup>	17 <sup>17.0%</sup>	17 <sup>15.5%</sup>	26 <sup>21.7%</sup>	21 <sup>16.2%</sup>	33 <sup>23.6%</sup>	35 <sup>23.3%</sup>
Layers: 6	12 <sup>20.0%</sup>	9 <sup>12.5%</sup>	26 <sup>31.0%</sup>	16 <sup>16.7%</sup>	24 <sup>22.2%</sup>	24 <sup>20.0%</sup>	25 <sup>18.9%</sup>	45 <sup>31.3%</sup>	54 <sup>34.6%</sup>	59 <sup>35.1%</sup>	55 <sup>30.6%</sup>
Layers: 7	11 <sup>15.7%</sup>	9 <sup>10.7%</sup>	10 <sup>10.2%</sup>	18 <sup>16.1%</sup>	17 <sup>13.5%</sup>	7 <sup>5.0%</sup>	8 <sup>5.2%</sup>	18 <sup>10.7%</sup>	21 <sup>11.5%</sup>	11 <sup>5.6%</sup>	32 <sup>15.2%</sup>
Layers: 8	5 <sup>6.3%</sup>	12 <sup>12.5%</sup>	5 <sup>4.5%</sup>	29 <sup>22.7%</sup>	14 <sup>9.7%</sup>	30 <sup>18.8%</sup>	57 <sup>32.4%</sup>	42 <sup>21.9%</sup>	19 <sup>9.1%</sup>	20 <sup>8.0%</sup>	27 <sup>11.3%</sup>
Layers: 9	8 <sup>8.9%</sup>	4 <sup>3.7%</sup>	22 <sup>17.5%</sup>	4 <sup>2.8%</sup>	16 <sup>9.9%</sup>	23 <sup>12.8%</sup>	44 <sup>22.2%</sup>	19 <sup>8.8%</sup>	25 <sup>10.7%</sup>	25 <sup>9.9%</sup>	20 <sup>7.4%</sup>
Layers: 10	5 <sup>5.0%</sup>	27 <sup>22.5%</sup>	11 <sup>7.9%</sup>	39 <sup>24.4%</sup>	30 <sup>16.7%</sup>	60 <sup>30.0%</sup>	26 <sup>11.8%</sup>	8 <sup>3.3%</sup>	7 <sup>2.7%</sup>	36 <sup>12.9%</sup>	38 <sup>12.7%</sup>
Layers: 11	48 <sup>43.6%</sup>	16 <sup>12.1%</sup>	34 <sup>22.1%</sup>	13 <sup>7.39%</sup>	7 <sup>3.5%</sup>	31 <sup>14.1%</sup>	21 <sup>8.7%</sup>	33 <sup>12.5%</sup>	58 <sup>20.3%</sup>	45 <sup>14.6%</sup>	41 <sup>12.4%</sup>
Layers: 12	18 <sup>15.0%</sup>	30 <sup>20.8%</sup>	15 <sup>8.9%</sup>	46 <sup>24.0%</sup>	58 <sup>26.9%</sup>	8 <sup>3.3%</sup>	13 <sup>4.9%</sup>	26 <sup>9.0%</sup>	41 <sup>13.1%</sup>	20 <sup>6.0%</sup>	16 <sup>4.4%</sup>
Layers: 13	13 <sup>10.0%</sup>	24 <sup>15.4%</sup>	41 <sup>22.5%</sup>	29 <sup>13.9%</sup>	38 <sup>16.2%</sup>	18 <sup>6.9%</sup>	58 <sup>20.3%</sup>	41 <sup>13.1%</sup>	42 <sup>12.4%</sup>	87 <sup>23.9%</sup>	29 <sup>7.4%</sup>
Layers: 14	7 <sup>5.0%</sup>	12 <sup>7.1%</sup>	49 <sup>25.0%</sup>	30 <sup>13.4%</sup>	4 <sup>1.6%</sup>	30 <sup>10.7%</sup>	71 <sup>23.1%</sup>	32 <sup>9.5%</sup>	48 <sup>13.2%</sup>	45 <sup>11.5%</sup>	75 <sup>17.9%</sup>
Layers: 15	35 <sup>23.3%</sup>	28 <sup>15.6%</sup>	38 <sup>18.1%</sup>	31 <sup>12.9%</sup>	30 <sup>11.1%</sup>	23 <sup>7.7%</sup>	29 <sup>8.8%</sup>	16 <sup>4.4%</sup>	30 <sup>7.7%</sup>	72 <sup>17.1%</sup>	36 <sup>8.0%</sup>
Threshold:80											
Layers: 5	7 <sup>(14.0%)</sup>	5 <sup>(8.3%)</sup>	9 <sup>(12.9%)</sup>	5 <sup>(6.3%)</sup>	15 <sup>(16.7%)</sup>	7 <sup>(7%)</sup>	12 <sup>(10.9%)</sup>	13 <sup>(10.8%)</sup>	10 <sup>(7.7%)</sup>	20 <sup>(14.3%)</sup>	16 <sup>(10.7%)</sup>
Layers: 6	4 <sup>(6.7%)</sup>	4 <sup>(5.6%)</sup>	15 <sup>(17.9%)</sup>	8 <sup>(8.3%)</sup>	10 <sup>(9.3%)</sup>	8 <sup>(6.7%)</sup>	12 <sup>(9.1%)</sup>	25 <sup>(17.3%)</sup>	25 <sup>(16.0%)</sup>	31 <sup>(18.5%)</sup>	27 <sup>(15.0%)</sup>
Layers: 7	5 <sup>(7.1%)</sup>	5 <sup>(6.0%)</sup>	4 <sup>(4.1%)</sup>	10 <sup>(8.9%)</sup>	9 <sup>(7.1%)</sup>	2 <sup>(1.4%)</sup>	6 <sup>(3.9%)</sup>	10 <sup>(6.0%)</sup>	7 <sup>(3.9%)</sup>	5 <sup>(2.6%)</sup>	19 <sup>(9.1%)</sup>
Layers: 8	4 <sup>(5.0%)</sup>	9 <sup>(9.4%)</sup>	4 <sup>(3.6%)</sup>	14 <sup>(10.9%)</sup>	7 <sup>(4.9%)</sup>	11 <sup>(6.9%)</sup>	22 <sup>(12.5%)</sup>	21 <sup>(10.9%)</sup>	8 <sup>(3.9%)</sup>	6 <sup>(2.7%)</sup>	8 <sup>(3.3%)</sup>
Layers: 9	2 <sup>(2.2%)</sup>	1 <sup>(0.9%)</sup>	10 <sup>(7.9%)</sup>	2 <sup>(1.4%)</sup>	8 <sup>(4.9%)</sup>	9 <sup>(5.0%)</sup>	21 <sup>(10.6%)</sup>	10 <sup>(4.6%)</sup>	11 <sup>(4.7%)</sup>	8 <sup>(3.2%)</sup>	10 <sup>(3.7%)</sup>
Layers: 10	3 <sup>(3%)</sup>	12 <sup>(10.0%)</sup>	7 <sup>(5.0%)</sup>	18 <sup>(11.3%)</sup>	11 <sup>(6.1%)</sup>	30 <sup>(15.0%)</sup>	8 <sup>(3.6%)</sup>	4 <sup>(1.7%)</sup>	6 <sup>(2.3%)</sup>	24 <sup>(8.6%)</sup>	18 <sup>(6.0%)</sup>
Layers: 11	29 <sup>(26.4%)</sup>	11 <sup>(8.3%)</sup>	14 <sup>(9.1%)</sup>	4 <sup>(2.3%)</sup>	2 <sup>(1.0%)</sup>	11 <sup>(5.0%)</sup>	5 <sup>(2.1%)</sup>	5 <sup>(1.9%)</sup>	5 <sup>(1.8%)</sup>	17 <sup>(5.5%)</sup>	18 <sup>(5.5%)</sup>
Layers: 12	13 <sup>(10.8%)</sup>	11 <sup>(7.6%)</sup>	6 <sup>(3.6%)</sup>	21 <sup>(10.9%)</sup>	26 <sup>(12.0%)</sup>	4 <sup>(1.7%)</sup>	12 <sup>(4.6%)</sup>	13 <sup>(4.5%)</sup>	10 <sup>(3.2%)</sup>	8 <sup>(2.4%)</sup>	4 <sup>(1.1%)</sup>
Layers: 13	8 <sup>(6.2%)</sup>	13 <sup>(8.3%)</sup>	21 <sup>(11.5%)</sup>	13 <sup>(6.3%)</sup>	14 <sup>(6.0%)</sup>	8 <sup>(3.1%)</sup>	23 <sup>(8.0%)</sup>	15 <sup>(4.8%)</sup>	20 <sup>(5.9%)</sup>	32 <sup>(8.8%)</sup>	12 <sup>(3.1%)</sup>
Layers: 14	4 <sup>(2.9%)</sup>	7 <sup>(4.2%)</sup>	22 <sup>(11.2%)</sup>	14 <sup>(6.3%)</sup>	4 <sup>(1.6%)</sup>	12 <sup>(4.3%)</sup>	41 <sup>(13.3%)</sup>	13 <sup>(3.9%)</sup>	9 <sup>(2.5%)</sup>	15 <sup>(3.8%)</sup>	24 <sup>(5.7%)</sup>
Layers: 15	15 <sup>(10.0%)</sup>	15 <sup>(8.3%)</sup>	15 <sup>(7.1%)</sup>	6 <sup>(2.5%)</sup>	15 <sup>(5.6%)</sup>	7 <sup>(2.3%)</sup>	13 <sup>(3.9%)</sup>	8 <sup>(2.2%)</sup>	9 <sup>(2.3%)</sup>	30 <sup>(7.1%)</sup>	9 <sup>(2.0%)</sup>

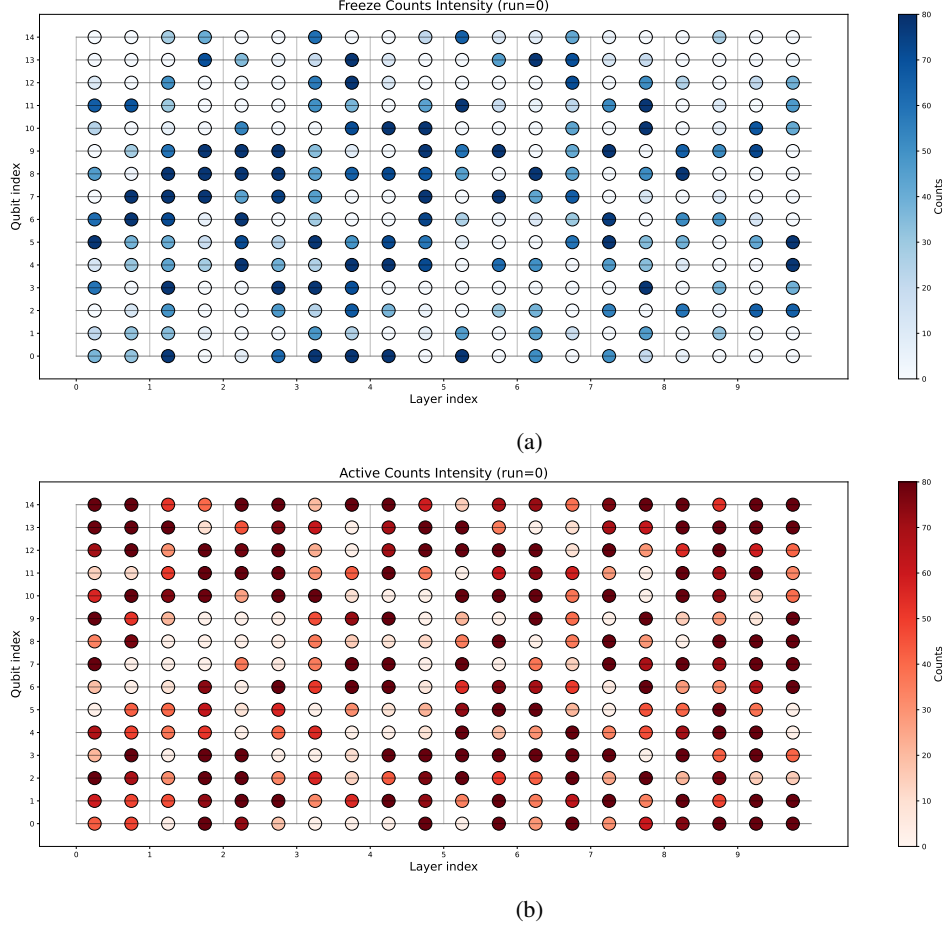
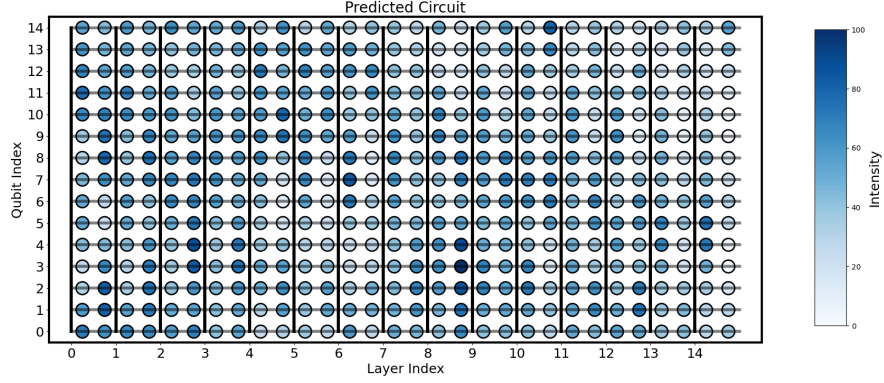
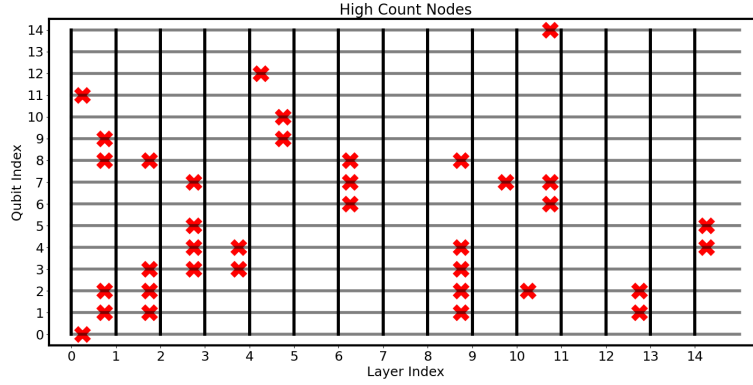


Figure 11: APFA records parameters trajectory with HEA-based isotropic Hamiltonian solver. (a) Freeze Counts Intensity: In the figure (a) above, the color of the dot indicates the number of times the parameter was "frozen" (freeze counts) during the optimization process. The darker the color, the more times the parameter was frozen; the lighter the color, the fewer times it was frozen. The thin black line on the outer edge of each dot is only used to emphasize the position of different parameters and has nothing to do with intensity. The color scale bar on the right indicates the value range and mapping relationship. (b) Active Counts Intensity: In the figure (b) below, the color of the dot indicates the number of times the parameter was "activated" (active counts) during the training process. Similarly, the darker the color, the more times it was activated; the lighter the color, the fewer times it was activated. Other marking methods are the same as in (a).

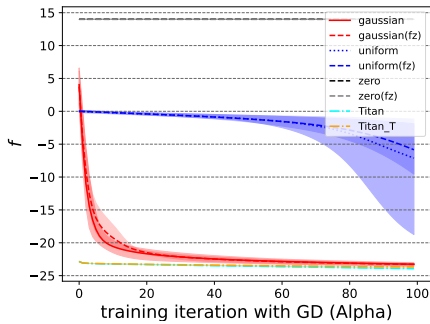


(a)

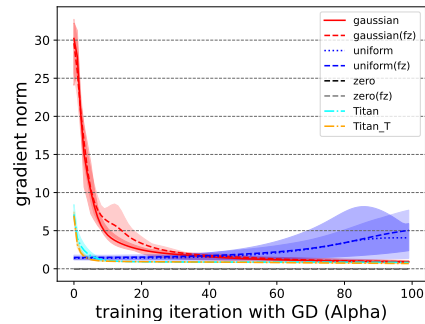


(b)

Figure 12: (a) TITAN predicted intensity. (b) TITAN selected Frozen Gates (**36/450**) (8%). Qubits 15, Layers 15, Threshold: 70. HEA-based isotropic Hamiltonian solver:  $XX + YY + ZZ$ .

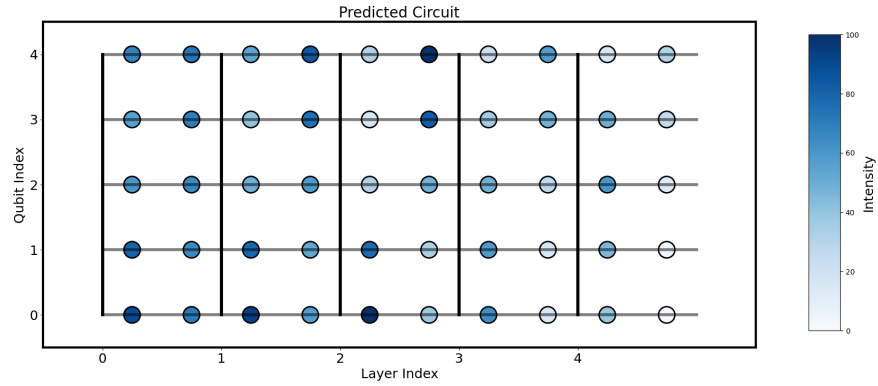


(a)

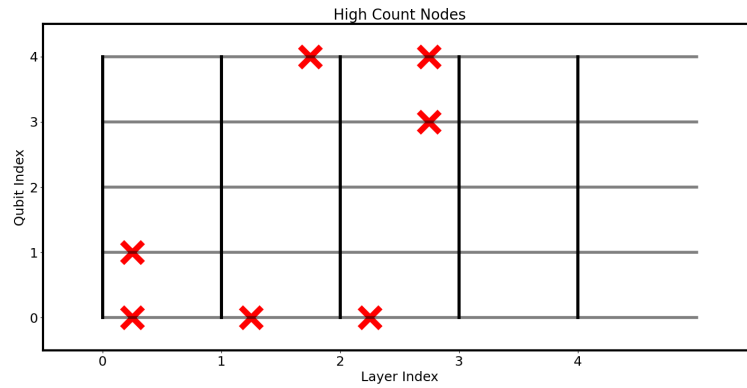


(b)

Figure 13: (a) Loss GD Noiseless. (b) Gradient Norm GD Noiseless. Qubits 15, Layers 15, Threshold: 70. Selected Frozen Gates (**36/450**) (8%). HEA-based isotropic Hamiltonian solver:  $XX + YY + ZZ$ . TITAN: Enhanced Gaussian. TITAN\_T: APFA Only without freezing.

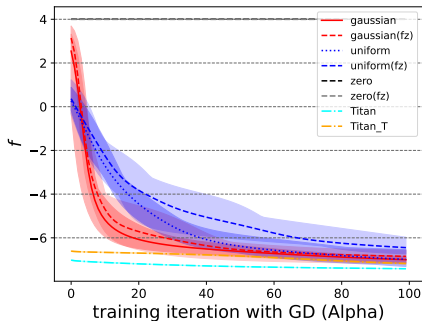


(a)

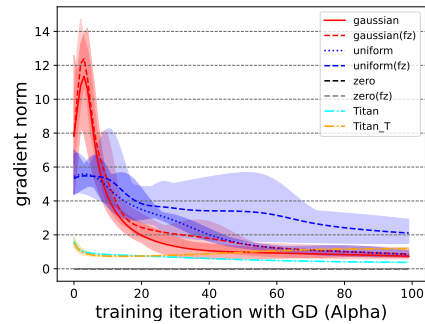


(b)

Figure 14: (a) TITAN predicted intensity. (b) TITAN selected Frozen Gates (**7/50**) (14%). Qubits 5, Layers 5, Threshold:  $\tau = 80$ . HEA-based isotropic Hamiltonian solver:  $XX + YY + ZZ$ .



(a)



(b)

Figure 15: (a) Loss GD Noiseless. (b) Gradient Norm GD Noiseless. Qubits 5, Layers 5, Threshold:  $\tau = 80$ . Selected Frozen Gates (**7/50**) (14%). HEA-based isotropic Hamiltonian solver:  $XX + YY + ZZ$ . TITAN: Enhanced Gaussian. TITAN\_T: APFA Only without freezing.

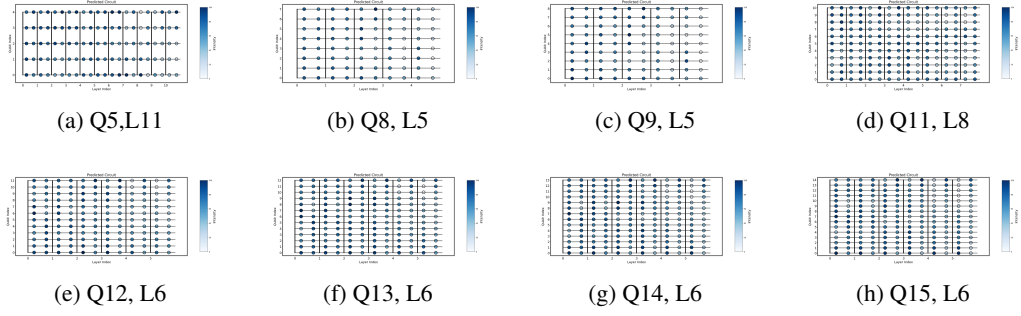


Figure 16: TITAN predicted freezing intensity for HEA-based isotropic Hamiltonian solver with threshold: 70.

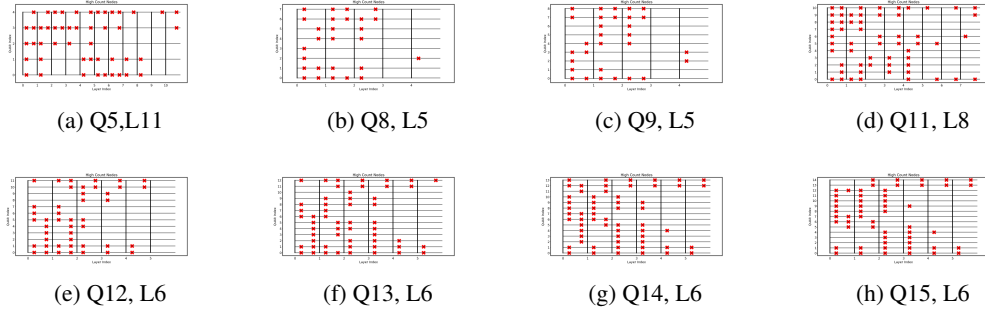


Figure 17: TITAN selected frozen Parameters for HEA-based isotropic Hamiltonian solver with threshold: 70.

Layers	Qubit											
	Qubit: 5	Qubit: 6	Qubit: 7	Qubit: 8	Qubit: 9	Qubit: 10	Qubit: 11	Qubit: 12	Qubit: 13	Qubit: 14	Qubit: 15	
Threshold:50												
Layers: 5	2652.0%	3355.0%	4260.0%	5568.8%	6066.7%	5050.0%	4843.6%	5243.3%	4736.2%	7050.0%	8858.7%	
Layers: 6	2541.7%	2636.1%	4654.8%	4647.9%	5651.9%	5445.0%	6045.5%	10774.3%	11171.2%	11870.2%	11765.0%	
Layers: 7	3144.3%	3744.1%	2929.6%	4035.7%	4535.7%	4431.4%	3422.1%	5733.9%	6736.8%	4824.5%	9746.2%	
Layers: 8	3442.5%	3435.4%	3934.8%	8264.1%	6142.4%	8150.6%	12269.3%	10152.6%	7837.5%	9642.9%	12853.3%	
Layers: 9	4044.4%	3229.6%	5846.0%	5135.4%	5936.4%	7943.9%	11457.6%	7132.9%	11147.4%	9738.5%	9334.4%	
Layers: 10	4040.0%	6655.0%	5035.7%	8351.9%	7843.3%	14874.0%	9342.3%	5121.3%	5119.6%	10537.5%	16354.3%	
Layers: 11	8476.4%	3929.6%	8555.2%	6235.2%	4020.2%	9643.6%	10342.6%	8130.7%	8028.0%	14547.1%	16550.0%	
Layers: 12	5142.5%	6947.9%	5532.7%	11057.3%	13160.7%	7129.6%	12447.0%	9834.0%	13342.6%	11534.2%	12534.7%	
Layers: 13	4937.7%	8051.3%	11965.4%	7737.0%	11850.4%	7930.4%	14751.4%	15449.4%	15044.4%	22261.0%	12532.1%	
Layers: 14	5841.4%	3822.6%	12061.2%	9241.1%	6827.0%	11139.6%	16854.6%	14242.3%	20857.1%	18747.7%	23155.0%	
Layers: 15	10368.7%	8848.9%	9645.7%	9037.5%	8732.2%	9632.0%	13942.1%	9726.9%	14436.9%	21150.2%	20645.8%	
Threshold:60												
Layers: 5	1836.0%	2135.0%	2840.0%	4151.3%	4246.7%	3333.0%	3330.0%	3630.0%	2720.8%	4431.4%	4832.0%	
Layers: 6	1626.7%	1926.4%	3642.9%	2728.1%	3936.1%	3831.7%	4433.3%	7652.8%	8453.8%	9254.8%	8647.8%	
Layers: 7	1927.1%	2125.0%	1818.4%	3026.8%	2923.0%	2316.4%	2013.0%	3520.8%	3619.8%	2211.2%	6028.6%	
Layers: 8	1518.8%	1818.8%	1513.4%	5442.2%	3121.5%	6037.5%	9554.0%	6533.9%	4320.7%	4520.1%	7230.0%	
Layers: 9	2426.7%	1513.9%	3628.6%	2013.9%	3219.8%	4927.2%	7638.4%	3415.7%	6025.6%	5120.2%	4817.8%	
Layers: 10	2222.0%	4840.0%	2417.1%	6540.6%	5329.4%	10854.0%	5223.6%	2912.1%	207.7%	6322.5%	8528.3%	
Layers: 11	7265.5%	2115.9%	6139.6%	3821.6%	2110.6%	5725.9%	5522.7%	3312.5%	2910.1%	8226.6%	9528.8%	
Layers: 12	3125.8%	5236.1%	3219.1%	8142.2%	9242.6%	2410.0%	8733.0%	4716.3%	7524.0%	5115.2%	5916.4%	
Layers: 13	2922.3%	4730.1%	8044.0%	4622.1%	8034.2%	3513.5%	10737.4%	8226.3%	9026.6%	14339.3%	5814.9%	
Layers: 14	2920.7%	2414.3%	8643.9%	6227.7%	228.7%	6121.8%	12039.0%	7923.5%	10629.1%	10727.3%	14033.3%	
Layers: 15	6744.7%	5228.9%	6330.0%	5824.2%	5520.4%	5919.7%	7623.0%	4412.2%	7619.5%	13532.1%	10623.6%	

Table 5: TITAN predicted frozen parameter with HEA-based isotropic Hamiltonian solver. Frozen parameters exceeding 30% is colored with red and exceeding between 10% and 30% is colored with blue and exceeding 60% is colored with darker red.

## F.4 Limitations

All empirical evidence is obtained on state–vector *classical* simulators. As a consequence, noise processes are injected only synthetically (Gaussian perturbations to gradients) rather than captured from *in situ* device characterization. The absence of crosstalk, SPAM errors, and non-Markovian decoherence. Scalability of APFA thresholds and EMA hyperparameters beyond this regime remains untested. This study remains limited to classical simulators and benchmark circuits do not exceed 100 qubits, however, in the future, we plan to scale to deeper circuits in real-world quantum sensors, ultimately enabling resource-efficient VQE deployments for chemically and physically relevant systems.

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